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Research Article

Fabrication and Characterization of Two-Dimensional Layered MoS_2 Thin Films by Pulsed Laser Deposition

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Direct growth of uniform wafer-scale two-dimensional (2D) layered materials using a universal method is of vital importance for utilizing 2D layers into practical applications. Here, we report on the structural and transport properties of large-scale few-layer MoS_2 back-gated field effect transistors (FETs), fabricated using conventional pulsed laser deposition (PLD) technique. Raman spectroscopy and transmission electron microscopy results confirmed that the obtained MoS_2 layers on SiO_2/Si substrate are multilayers. The FETs devices exhibit a relative high on/off ratio of 5×10^2 and mobility of $0.124\,cm^2V^{-1}S^{-1}$. Our results suggest that the PLD would be a suitable pathway to grow 2D layers for future industrial device applications.

1. Introduction

Atomically layered two-dimensional (2D) materials are intriguing for both electronic and optoelectronic applications, because of their unique electrical, optical, and mechanical properties [1–4]. Among these 2D layered materials, molybdenum dichalcogenides (MoS_2), where the layered S–Mo–S are bonded by van der Waals interactions with Mo and S atoms which are bonded by strong covalent interactions, have attracted extensive interests due to their considerable band gap and high carrier mobility. Both theoretical estimations and experimental observations indicate an indirect to direct energy bandgap transition when MoS_2 is thinned from bulk to monolayer, while its bandgap increases from ~1.2 eV to ~1.8 eV [5–8]. Owing to the extraordinary layer-dependent bandgap behavior, MoS_2 is considered a promising candidate to overcome the shortages belonging to zero-bandgap graphene, providing a possible solution for next-generation electronic applications [9]. For instance, monolayer and few-layer MoS_2 based field effect transistors (FETs) have been reported, possessing high on/off ratios exceeding 10^3 [10–12]. The mobility in monolayer MoS_2 FETs in vacuum reported so far is $8\,cm^2v^{-1}s^{-1}$ [13–15], which is lower than the theoretical value ($410\,cm^2v^{-1}s^{-1}$) [15]. The mobility in few-layer FETs at room temperature could reach $470\,cm^2v^{-1}s^{-1}$[16]; the value is close to the theoretical value ($200{\sim}500\,cm^2v^{-1}s^{-1}$) estimated through phonon scattering limit [17]. Even with the desirable requirement for industrial applications, to date, most of works have focused on exploring the methods of large scale, high quality MoS_2 thin films. Conventional methods to obtain MoS_2 layers include mechanical exfoliation, chemical vapor deposition (CVD), and PLD [18–20]. However, it is inconvenient to control the scale size and thickness of samples using mechanical exfoliation method. The layer nucleation in CVD methods is difficult with a rigor experimental condition, resulting nonuniform MoS_2 layers. Up to now, PLD method has successfully been employed for the deposition of large-scale layered materials, including graphene, black phosphorous (BP), MoS_2, and InSe [21–23]. One of the main advantages of using PLD is the convenience to control the layer numbers of the large-scale 2D materials by solely varying the pulses numbers. In this work, we report the structural and transport characterization of few-layer MoS_2 back-gated FETs fabricated via PLD technique.

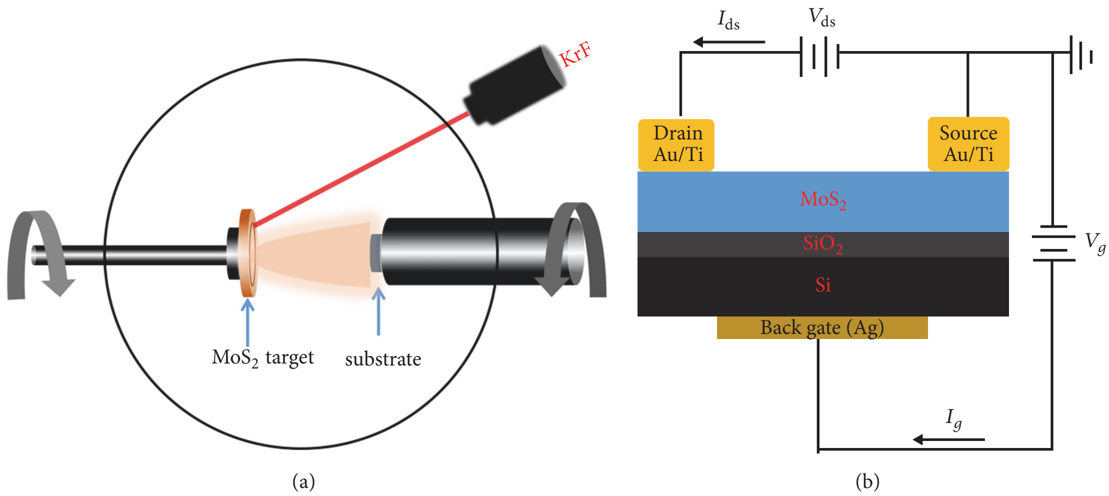


Figure 1: Schematic diagram of (a) PLD equipment; (b) MoS_2 back-gated FETs structure.

2. Materials and Methods

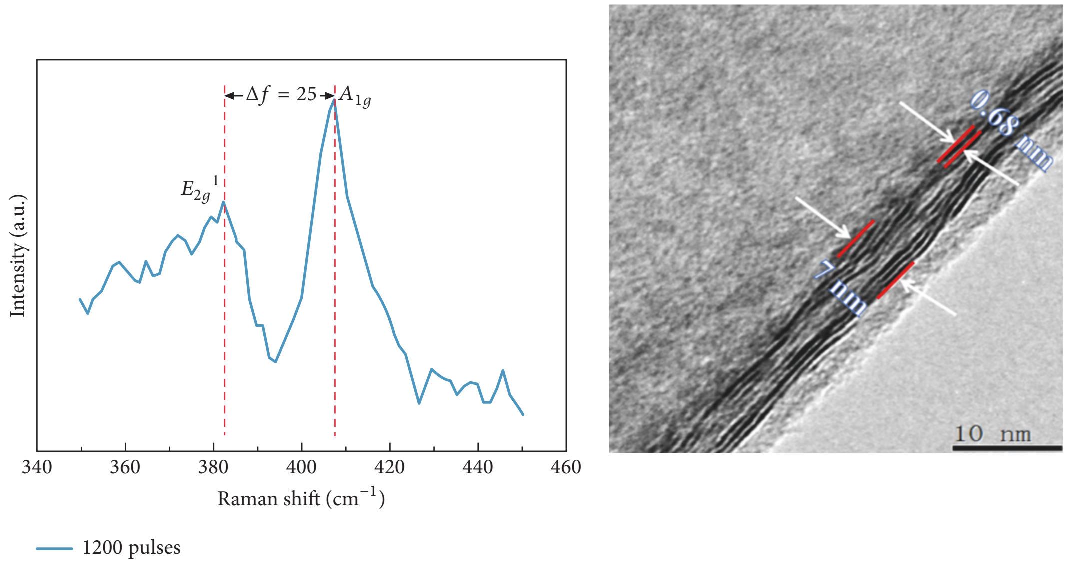
Few-layer MoS_2 films with the dimension of 10 mm × 10 mm were fabricated on the surface-cleaned SiO_2 (300 nm)/p$^+$-Si wafer by PLD (KrF 248 nm) with a laser energy of 200 mJ/pulses and a frequency of 5 Hz. Figure 1(a) illustrates the experimental setup of the PLD system. A commercial MoS_2 pellet (HeFei Crystal Technical Material Co., Ltd.) was used as the target. The target and substrate distance was set at 5 cm. Before deposition, the chamber was evacuated to a base pressure of ~1×10^{-5} Pa to avoid the oxidation of the MoS_2 films. The SiO_2/Si substrates temperature was maintained at 700°C during the growth. Both target and substrate were rotated during the deposition to obtain a uniform film. When the laser pulses strike the MoS_2 target, the formed plasma including atoms and ions will reach substrates. The thickness of MoS_2 films can be controlled by the number of laser pulses. Here the pulse numbers for growth MoS_2 films are set to be 1200 pulses. The atoms valence in MoS_2 was measured by X-ray photoelectron spectroscopy (XPS). Raman spectroscopy and transmission electron microscopy (TEM) were employed to confirm the thickness of MoS_2 films. To construct the FETs structure, radio frequency magnetron sputtering technique was used to deposit Au/Ti electrodes on surface of MoS_2 layers and back of SiO_2/Si substrates. The deposition time of Au/Ti electrodes is 2 min and 20 s, respectively. The schematic diagram of the fabricated FETs device is shown in Figure 1(b). The transport and transfer measurements were conducted in a Keithley 4200 semiconductor characterization system. All the tests were executed at 300 K in atmosphere.

3. Results and Discussion

Raman spectroscopy was a commonly used technique to estimate the thickness of the 2D materials [24]. As shown in Figure 2(a), two prominent characteristic peaks are attributed to the mode of MoS_2 atomic layers vibration (in-plane ${E_{2g}}^1$ mode: ~382 cm^{-1} and out-of-plane A_{1g} mode: ~407 cm^{-1}). The distance between ${E_{2g}}^1$ and A_{1g} Δf is proportional to thickness of MoS_2 films, the ${E_{2g}}^1$ peak exhibits a red shift with a blue shift of the A_{1g} peak when the MoS_2 thickness decreased, and Δf follows an empirical formula: $\Delta f = 26.45 - 15.42/(1 + 1.44n^{0.9})$ cm^{-1} [25]. According to the formula, the thickness of MoS_2 layers can be deduced: monolayer ~18 cm^{-1}, bilayer ~22.4 cm^{-1}, trilayer ~23 cm^{-1}, and few-layer ~25 cm^{-1}. The cross-sectional TEM image (shown in Figure 2(b)) exhibits a sharp and well-defined MoS_2 layers. A 7 nm thick MoS_2 layer was grown on SiO_2/Si wafer with an interlayer spacing ~0.68 nm, which is close to the theoretical value of the MoS_2 monolayer thickness (~0.65 nm) [26]. These results are consistent with the Raman spectral results, implying that the obtained MoS_2 films ($\Delta f = 25$ cm^{-1}) in this work are few-layer sample.

In order to examine the sulfur deficiency in the MoS_2 films, XPS analysis was carried out. As shown in Figure 3, there are three peaks of Mo 3d, located at 229.18 eV, 232.67 eV, and 235.97 eV. The first two peaks could be assigned to Mo $3d_{5/2}$ and Mo $3d_{3/2}$, respectively. They are correlating to Mo^{4+} state in MoS_2. The third core level peak is correlating to Mo^{6+} $3d_{3/2}$ state, and the fitted curves are shown in blue in the image. Due to the existence of Mo^{6+}, there should be S vacancies in our samples. One possibility is the S missing during the process of the growth of the films. Other possibility is the reaction with oxygen to form MoO_3 after the sample taking out of the chamber. The existence of MoO_3 will exert influence on the optical and electrical characteristics of MoS_2 thin films.

To investigate the electric properties of the MoS_2 film, back-gated FETs were fabricated based on few-layer MoS_2 films grown on SiO_2/Si substrates. As seen from Figure 4(a), the relationship between drain current (I_{ds}) and drain voltage (V_{ds}) at different back-gated voltage (V_g) was investigated, and the step of back-gated voltage is 5 V. A clear n-type semiconducting property was depicted which is consistent with previous reports [27]. The n-type behavior of MoS_2 sample might be due to the impurities or existence of interstitial



(a) (b)

FIGURE 2: (a) Raman image of MoS_2 films. The distance between $E_{2g}{}^{1}$ and A_{1g} is 25 cm^{-1}. (b) Cross-sectional TEM image of few-layer MoS_2 film.

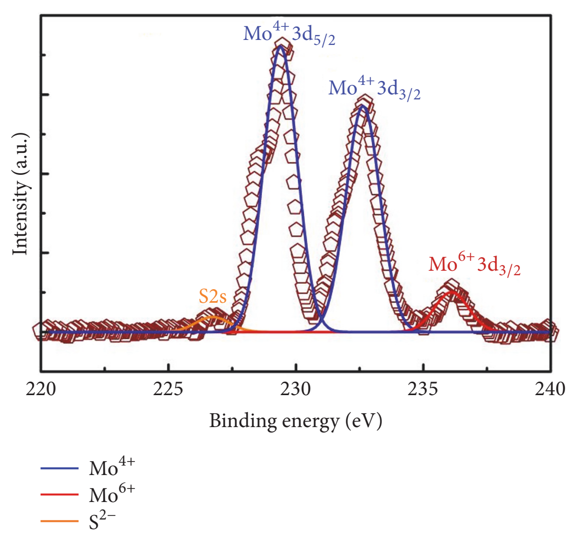


FIGURE 3: The XPS spectra of Mo 3d, S 2s core levels in MoS_2 films.

atoms in the interlayer gap. As shown in Figure 4(b), the off-state leakage current (I_{off}) is ~10 nA, while the on-state current (I_{on}) is ~5 μA; the on/off ratio is about 5×10^2, and the ratio is compared to the ratio (~10^3) of monolayer FETs [10]. The mobility of multilayer MoS_2 FETs can be estimated based on the formula:

$$\mu = \frac{L}{W \times (\varepsilon_0 \varepsilon_r / d) \times V_{\text{ds}}} \times \frac{dI_{\text{ds}}}{dV_g} \tag{1}$$

and the channel length L is 0.1 mm, the channel width W is 1.5 mm, ε_0 is 8.854×10^{-12} F/m, ε_r is 3.9 [17], d is the thickness of SiO_2 (300 nm), and V_{ds} in our experiment is 5 V. The value of dI_{ds}/dV_g can be obtained from the linear fit in Figure 4(b). The mobility of the FETs calculated is 0.124 $cm^2v^{-1}s^{-1}$, and the value is similar to other multilayer back-gated MoS_2 FETs [28]. A bit low mobility may be due to the impurity phase in the film or the impurities and traps on the surface of SiO_2 films [17]. Moreover, the nonlinear rectifying behavior in I_{ds}-V_{ds} curves and the saturation of current in the high positive gate voltage imply a contact resistance between the electrodes and the film, which could also result in an underestimated mobility.

4. Conclusions

In summary, we have reported the PLD grown few-layer MoS_2 based FETs showing good transport characteristics

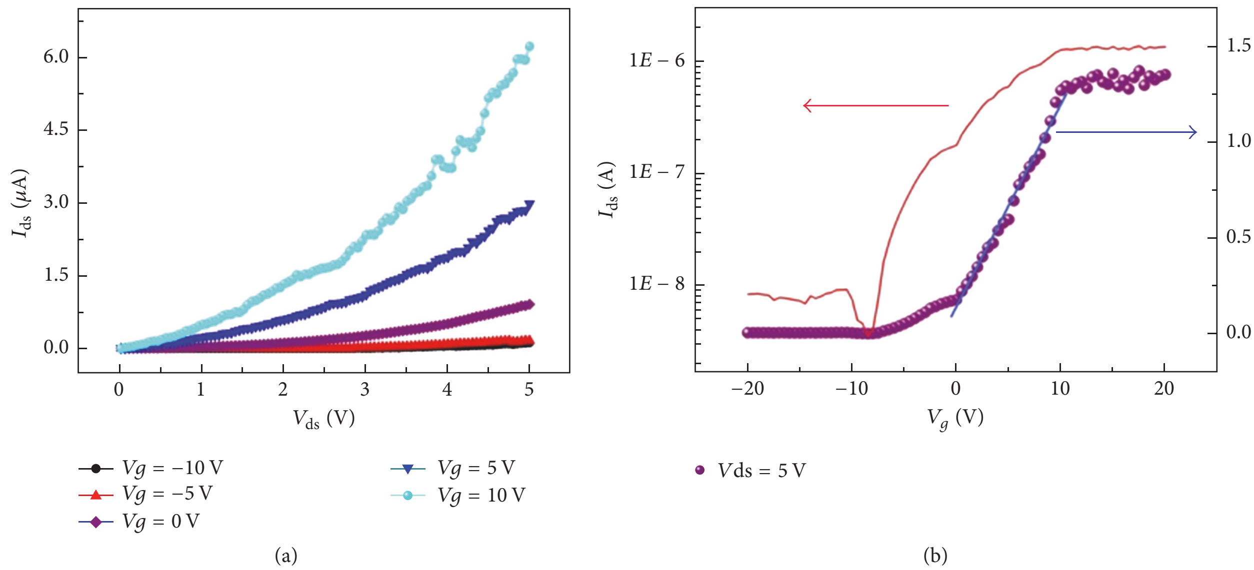


Figure 4: FETs characterization of few-layer MoS_2 film. (a) Transport characteristics at different V_g. (b) Transfer curves of MoS_2 thin films in logarithmic and linear scales.

with relatively high ON/OFF ratio ~500, which is comparable to monolayer and few-layer MoS_2 FETs fabricated using CVD-grown samples. Our results suggest that the PLD grown MoS_2 films not only achieve large-scale size, but also present moderate electric properties. The developed PLD method for growth of wafer-scale 2D layered materials may provide a new insight for further electronic applications.

Conflicts of Interest

The authors declare no conflicts of interest.

Acknowledgments

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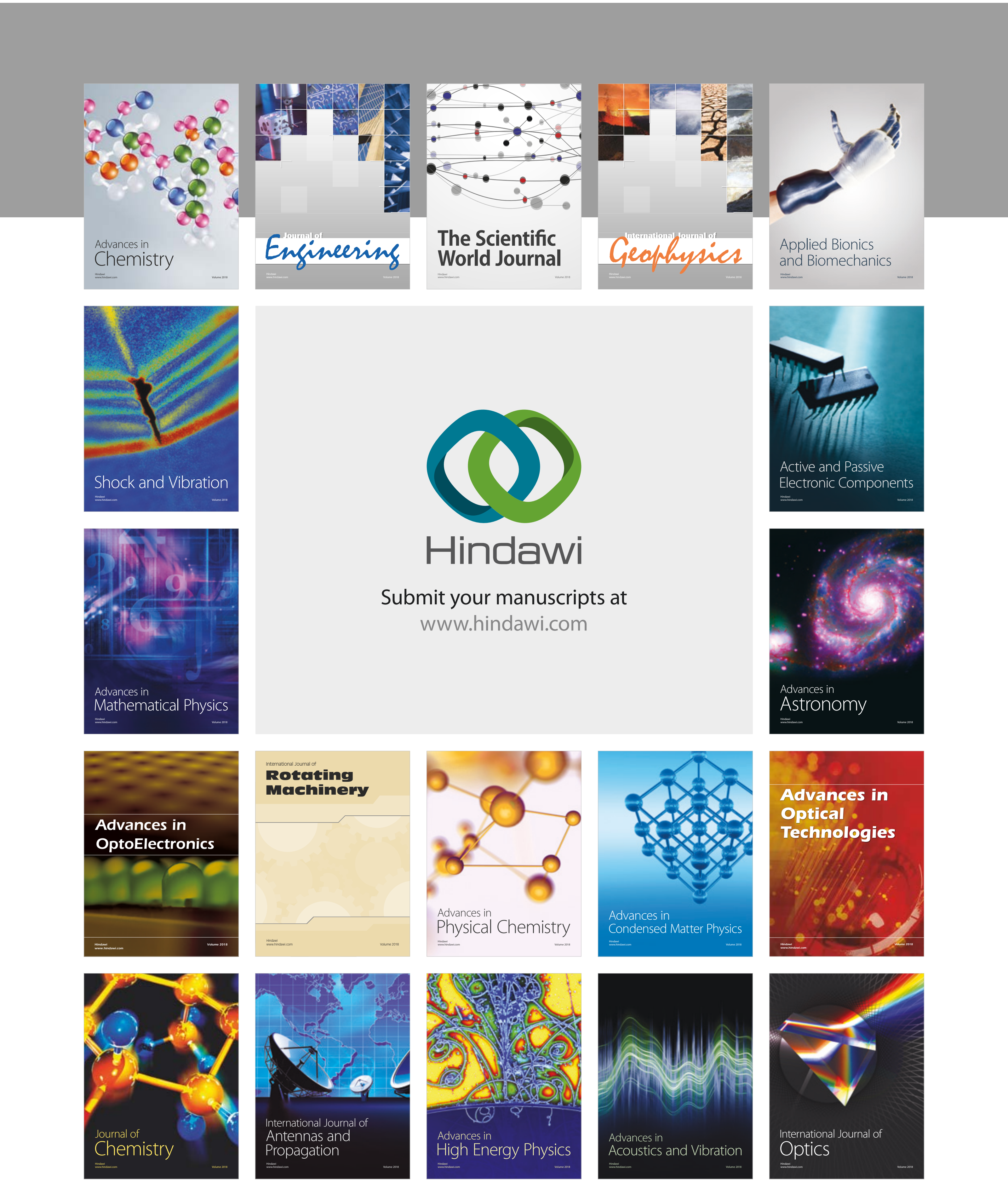
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