Low-Temperature Chemical Vapor Deposition Growth of MoS2 Nanodots and Their Raman and Photoluminescence Profiles

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We report on the growth of an ordered array of MoS2 nanodots (lateral sizes in the range of ~100–250 nm) by a thermal chemical vapor deposition (CVD) method directly onto SiO2 substrates at a relatively low substrate temperature (510–560°C). The temperature-dependent growth and evolution of MoS2 nanodots and the local environment of sulfur-induced structural defects and impurities were systematically investigated by field emission scanning electron microscopy, micro-Raman spectroscopy, and X-ray photoelectron spectroscopy (XPS) techniques. At the substrate temperature of 560°C, we observed mostly few-layer MoS2, and at 510°C, multilayer MoS2 growth, as confirmed from the Raman line shape analysis. With reduced substrate temperature, the density of MoS2 nanodots decreases, and layer thickness increases. Raman studies show characteristic Raman modes of the crystalline MoS2 layer, along with two new Raman modes centered at ~346 and ~361 cm⁻¹, which are associated with MoO2 and MoO3 phases, respectively. Room temperature photoluminescence (PL) studies revealed strong visible PL from MoS2 layers, which is strongly blue-shifted from the bulk MoS2 flakes. The strong visible emission centered at ~658 nm signifies a free excitonic transition in the direct gap of single-layer MoS2. Position-dependent PL profiles show excellent uniformity of the MoS2 layers for samples grown at 540 and 560°C. These results are significant for the low-temperature CVD growth of a few-layer MoS2 dots with direct bandgap photoluminescence on a flexible substrate.

Keywords: MoS2 dots, photoluminescence, low temperature CVD, defect engineering, transition metal dichalcogenides

INTRODUCTION

Since the discovery of graphene, two-dimensional materials have attracted much interest due to their interesting optical, electrical, and mechanical characteristics. Molybdenum disulfide (MoS2), a transition metal dichalcogenide (TMD), has a layered structure of hexagonally arranged covalently bonded Mo and S atoms. The vast difference in the electronic structure of the bulk in comparison with the monolayer MoS2 offers a great opportunity for diverse applications. It has an excellent on/off ratio of 108 (Radisavljevic et al., 2011) and a high carrier mobility up to 200 cm²/Vs at room temperature (Radisavljevic et al., 2011), making it a promising candidate for optoelectronic applications, such as photodetectors (Yin et al., 2012) and solar cells (Lee et al., 2012) photovoltaics, (Feng et al.,
MoS2 dots using position-dependent micro-Raman spectroscopy and X-ray photoelectron spectroscopy (XPS) analyses. Some new Raman bands at 346 and 365 cm\(^{-1}\) were identified, besides the characteristic Raman signatures of MoS2 (E\(_{2g}\) and A\(_{1g}\)), which are associated with the presence of MoO2 and MoO3. The Raman features of the as-grown MoS2 dots were fully consistent with the XPS analysis. The XPS spectral line shape analysis of Mo-3d and S-2p core level peaks reveals a high amount of S vacancies in the as-grown MoS2 dots.

**EXPERIMENTAL**

Few layered MoS2 dots were directly grown on the SiO2 substrates using an in-house developed CVD system. Commercially procured MoO3 and S powder were used as precursors for the CVD growth of MoS2 in a quartz tube-based horizontal muffle furnace. High-purity argon (Ar) gas was used as the carrier gas. At first, the S precursor was kept near the entrance of the quartz tube, away from the central heating zone of the furnace, while the MoO3 powder was placed at the center of the furnace, and a few pieces of bare SiO2 substrates were placed near the source powder at the rear end side of the quartz tube. Note that both S and MoO3 precursors were separately loaded in two mini quartz tubes (inner diameter~10 mm) and kept at a distance of 53 and 33 cm away from the substrates (see Supplementary Figure S1). This arrangement provides stable evaporation of S and MoO3 sources at suitable temperatures and avoids any cross-contaminations during the MoS2 growth. The quartz chamber was pumped down to a base pressure of 1.9 \(\times\) 10\(^{-2}\) mbar, and the central zone temperature was raised to 760°C at the rate of 17°C/min for 45 min and was allowed to stabilize at this temperature for 15 min with Ar as the carrier gas with a flow rate of 100 sccm (standard cubic centimeters per minute), keeping the pressure at 2.2 mbar. Then, it was ramped up slowly to 800°C at the rate of 4°C/min and then, maintained at this temperature for 30 min without changing the gas flow. After the growth process, the system was allowed to cool down to room temperature with an Ar gas flow of 300 sccm. The experimental setup and the complete timeline profile of the CVD-grown MoS2 dots are shown in Supplementary Figure S1. The substrate temperature is controlled by the proper positioning of the substrate inside the furnace. The temperature at each position corresponding to the substrate was calibrated using an external thermocouple prior to the growth of MoS2 nanodots. The MoS2 growth was monitored at four different substrate temperatures. The samples grown at substrate temperatures of 560, 540, 510, and 310°C are named as MS-560, MS-540, MS-510, and MS-310, respectively.

**CHARACTERIZATION**

Micro-Raman and PL measurements were performed with a high-resolution spectrometer (Horiba 171 LabRam HR), with excitation wavelengths (\(\lambda_{ex}\)) of 488 and 514.5 nm (Ar+ ion laser). The excitation source was focused with a 100× objective lens, a
spot size of 2 μm, and a laser power of 1.5 mW, and the signal was collected by a CCD in a back scattering geometry sent through a multimode fiber grating of 1800 grooves/mm. X-ray photoelectron spectroscopy (XPS) measurements were performed with a fully automated XPS microprobe (PHI-Xtool, Ulvac-Phi) using an Al Kα X-ray beam (1486.7 eV). The standard carbon 1s spectrum was used to calibrate the XPS spectra recorded for various samples with the spectral resolution of 0.2 eV. Surface morphology and microstructure of the MoS₂ dots were examined by using a field emission scanning electron microscope (FESEM), FEI-Quanta 400 with an acceleration voltage of 20 kV with spatial resolution: 1.2 nm at 30 kV in SE, 2 nm at 30 kV in BSE (gold/carbon), detectors: SE, BSE. Spectral line shapes of the micro-Raman features were fitted by Lorentzian line shapes. The XPS line shape profile for all the samples was performed by Shirley peak fitting using Peakfit software with peak parameters as free variables. The best fit was obtained by a large number of iterations.

RESULTS AND DISCUSSIONS

FESEM Studies

In order to understand the morphology of the as-grown samples, FESEM imaging was carried out. A detailed description of the sample growth conditions and FESEM features is provided in Supplementary Table S1. Figures 1A,B show the FESEM images of sample MS-560 at two different magnifications, while Figures 1C,D represent the FESEM images of sample MS-510. It is clear that in each sample, MoS₂ dots of a variety of sizes are formed. The uniformity of these dots is almost maintained throughout the surface of the sample, estimated as 1 cm × 1 cm. We observed that the MS-560 has a doughnut-shaped structure of MoS₂ dots, which are denser and smaller than that of MS-510. The shape of the MS-510 dots is mostly circular, and a regular array type structure is formed. The observed regular array of MoS₂ dots is likely to be due to the direction of the outgoing carrier gas flow.
**AFM Studies**

Figures 2A,B show the AFM images of the as-grown samples grown at different substrate temperatures. For the same growth duration, but at different growth temperatures, we notice an evolution of the morphology of the MoS$_2$ nanodots. In the experiment, we observed that as the substrate is at higher temperature and close to the center of the quartz tube, higher density of the MoS$_2$ nanodots are formed and the corresponding sizes are smaller. In Figures 2A,B for the MS-560 sample, the MoS$_2$ nanodots show some kind of a ring-like structure. These rings are evenly distributed throughout the sample. The average size is estimated to be $\sim$200 nm. We can also see from the AFM height profile (Figure 2C) that bilayer and a few-layer MoS$_2$ were grown at this temperature (560°C). In Figures 2D,E, for the MS-540 sample, we notice that the MoS$_2$ ring structures are less in density than that of MS-560, and there is also presence of larger particle sizes ranging from $\sim$200 to 500 nm. These MoS$_2$ layers are found to be thicker, and from the height profile (Figure 2F), we can see that they are multilayered. Few larger circular ring structures are also present with diameter $\sim$1 µm. In Figures 2G,H, for MS-510 sample, the nanodots are seen to be less dense than that of MS-560 and MS-540 cases. There is also presence of larger circular rings with sizes ranging from $\sim$800 nm to 2 µm.
FIGURE 3 | (A) Raman fingerprints of as-grown MoS₂ dots grown at different substrate temperatures. (B) Lorentzian line shape fitting of E₂g and A₁g modes in MS-510. Note that the peak center of each mode is denoted in cm⁻¹ unit. (C) Position-dependent Raman spectra of the MS-560 sample, and (D) the corresponding optical micrograph image showing the scanned region for each spectrum marked with the respective serial number. (E) Lorentzian line shape fitting of E₂g and A₁g modes in MS-560 (position 1) along with the additional Raman modes. These spectra were recorded with 488 nm excitation.
the A1g and E2g. The observed

\[ \text{MS-310: 379.4 ± 0.1 \text{ cm}^{-1}} \]
\[ \text{MS-540: 387.3 ± 0.1 \text{ cm}^{-1}} \]
\[ \text{MS-560: 387.7 ± 0.1 \text{ cm}^{-1}} \]
\[ \text{MS-510: 409.0 ± 0.1 \text{ cm}^{-1}} \]
\[ \text{MS-310: 379.4 ± 0.1 \text{ cm}^{-1}} \]

few-layer MoS2, associated with the E2g and A1g phonon characteristic Raman modes, and this is a signature of the spectra for different samples, which exhibit two important

Asymmetric towards a higher wavenumber. (Li et al., 2012b). The as-grown MoS2 dot array in MS-560 and the

MS-510
\[ \text{409.4 \text{ cm}^{-1}} \]
\[ \text{379.4 \text{ cm}^{-1}} \]

MoS2 dot samples, and it was compared with that of the bulk MoS2 flakes. Figure 3A represents a comparison of the Raman spectra for different samples, which exhibit two important characteristic Raman modes, and this is a signature of the few-layer MoS2, associated with the E2g and A1g phonon modes at 379 and 405 cm\(^{-1}\), respectively. In general, the E2g mode is an in-plane vibration of two S atoms with respect to the Mo atoms, while the A1g mode is associated with the out-of-plane vibration of only S atoms in opposite directions with respect to the Mo atoms. These two Raman modes are very sensitive to the local in-plane, edge defects, and impurities present on the atomic layers of MoS2. The separation between these two modes is a measure of the number of layers in the MoS2. It is clear from Figure 3A that a few-layer MoS2 has been grown for samples MS-510, MS-540, and MS-510 as evidenced by the presence of E2g and A1g Raman bands, which is also consistent with the FESEM images (Figures 1A–D). On the other hand, in the case of sample MS-310, there was no signature of the MoS2 layer, as evidenced by the absence of the A1g peak in the Raman spectrum. The Raman spectrum of MS-310 shows a strong Raman peak at 338 cm\(^{-1}\), which signifies the presence of MoO3 and negligible presence of S.

We also observed a phonon mode stiffening of the characteristic Raman modes, reflected by a blue shift of both the peaks by 3–5 cm\(^{-1}\) for all the as-grown samples. The blue shift is expected to be closely related to the compressive strain induced by the adlayer-substrate lattice mismatch (Hui et al., 2013). The line shape parameters of Raman peaks are shown in Table 1, which depicts the peak position and frequency difference (\(\Delta k\)) between the A1g and E2g. The observed \(\Delta k\) values signify the growth of bilayer and trilayer MoS2. The Lorentzian line shape was fitted for the E2g and A1g bands, as shown in Figure 3B. There are two new Raman bands fitted at the lower and higher frequency side at 380 and 415 cm\(^{-1}\), respectively. An earlier report suggests that the splitting of the A1g peak into two peaks is due to the appearance of a Raman inactive mode (B1g) at 415 cm\(^{-1}\), which is not prominent in the case of bulk MoS2. However, it becomes significant in the case of monolayer MoS2, which makes the A1g peak (406.9 cm\(^{-1}\)) asymmetric towards a higher wavenumber. (Li et al., 2012b). The observed splitting in the E2g Raman mode at 380 cm\(^{-1}\) can be attributed to the mechanical strain in the MoS2 dots. (Conley et al., 2013). Note that the symbols represent the experimental data, and blue solid lines signify the fitted data in Figures 3B,E.

Figures 3C,D show the position-dependent Raman spectra of the as-grown MoS2 dot array in MS-560 and the corresponding OM image showing the scanned area for each spectrum marked with the respective serial number. We observed that the Raman spectra for different locations of a sample show quite similar features. In spots 1 and 2, we found that the frequency difference \(\Delta k\) between the Raman bands is ∼21.5 cm\(^{-1}\) and 21.9 cm\(^{-1}\), respectively, which implies the presence of bilayer MoS2. However, at spots 3 and 4, the frequency differences \(\Delta k\) are found to be 20.2 and 19.9 cm\(^{-1}\), respectively, which implies the coverage of few layers to monolayer MoS2. We notice a shift of the E2g mode without any significant shift of the A1g mode for different spots of the sample MS-560. The summary of the peak parameters is shown in Table 2. There are some additional Raman bands at 346.3, 351.6, and 365.3 cm\(^{-1}\), which are assigned to the stretching modes of the doubly coordinated oxygen (Mo=O–O–Mo). The observed Raman bands of MoO3 and MoO2 compare well with the reported literature values. (Dieterle and Mestl, 2002). Note that MoO2 and MoO3 are present in the sample due to low-temperature growth.

Position-dependent Raman spectra of as-grown MoS2 dots in MS-540 show similar uniformity and growth of monolayer and bilayer MoS2 dots, as shown in Supplementary Figure S2; Supplementary Table S2. There is also the presence of an additional band corresponding to the MoO2 and MoO3 phases (see Supplementary Figure S2; Supplementary Table S2). In the case of MS-510, we observed mostly trilayer and multi-layered MoS2 growth (Supplementary Figure S2; Supplementary Table S3). In this case, the E2g mode is downshifted substantially. Thus, growth at 540–560°C is suitable for the growth of monolayer and bilayer MoS2.

However, there are recent research works that have been progressing on low temperature CVD growth in which a new root is being proposed using the atmospheric CVD technique to grow monolayers of MoS2 at 550°C by using sodium (Na) as the catalyst. (Saefer et al., 2021). Authors have produced single and polycrystalline films by controlling the NaNO3/MoO3 catalyst/precursor ratio and growth time. Based on the first-principle calculations, authors have determined that Na was the nucleation site for the growth of MoS2, and the precursor’s ratio is crucial to decrease the formation energy and growth temperature. In the present case, the additional Raman bands associated with the E2g mode; all the samples are attributed to the nucleation sites of MoO3/ MoO2 that may be more favorable for the sulfurization process in order to form MoS2 nanodots at low-temperature regimes i.e., 510, 540, and 560°C.
XPS Studies

Figure 4 shows the core level XPS spectra of the Mo-3d and S-2p bands for the as-grown MoS$_2$ dots grown at different substrate temperatures. In Figures 4A,B, several Mo 3d$_{5/2}$ peaks could be fitted for each Mo (3d) envelope, indicating that more than one Mo species was present. In most of the spectra, the main Mo component (Mo$^{4+}$) was MoS$_2$. Besides MoS$_2$, all the samples contain additional Mo (3d) peaks corresponding to the oxides of the Mo metal (Mo$^{6+}$). Note that the peak positions and local environment of S induced surface defects, and impurities were estimated from the Gaussian line shape of the Mo (3d) and S (2p) core level peaks, and the fitting parameters along with the peak

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**TABLE 2** | $E_{2g}$ and $A_{1g}$ Raman modes at different locations of MS-560 and their peak separation significant for the growth of monolayer and bi-layer MoS$_2$.

| Sample location | $E_{2g}$ (cm$^{-1}$) | $A_{1g}$ (cm$^{-1}$) | $\Delta k$ (cm$^{-1}$) | Additional bands (cm$^{-1}$) | Error bar (cm$^{-1}$) |
|-----------------|---------------------|---------------------|----------------------|-----------------------------|----------------------|
| 1               | 385.4               | 406.9               | 21.5                 | 346.3, 351.6, 365.3          | ±0.1                 |
| 2               | 385.0               | 406.9               | 21.9                 | 345.5, 351.8, 364.9          | ±0.1                 |
| 3               | 387.5               | 407.7               | 20.2                 | –                            | ±0.1                 |
| 4               | 388.5               | 408.0               | 19.7                 | –                            | ±0.1                 |

**Identity**

| In plane vibration | Out of plane vibration | Few layer MoS$_2$ | Presence of MoO$_3$ and MoO$_2$ |

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**FIGURE 4** | Core level XPS spectra and the Shirley line shape fitting of: (A–C) Mo-3d (3d$_{5/2}$, 3d$_{3/2}$), (D–F) S-2p (2p$_{3/2}$, 2p$_{1/2}$) and (G–I) O-1s for MoS$_2$ dots grown at different substrate temperatures. The peak center in each case is denoted in the eV unit.
Identity are summarized in Table 3. Note that the as-grown MS-560 and MS-510 show quite similar spectra. However, in the case of MS-310 (Figure 4C), we notice that the peak intensity corresponding to MoO3 is much higher than that corresponding to MoS2, and the sulfur (2s) peak is absent. This may be due to the higher content of the physisorbed oxygen. Note that MS-310 is grown at a very low temperature, and due to the low vapor pressure of S, the formation of the MoO3 phase dominates.

Figures 4D–F shows the core level XPS S-2p spectra for the as-grown MoS2 dots. The band that is observed in S-2p is the 2p3/2, which is significant for the crystalline S bonded with Mo. The S-2p spectra for all the as-grown samples show the presence of a number of S components. The content of S in all the samples is found to be very low. For MS-310, the amount of S is found to be much less than that of MS-560 and MS-510. This may be due to the lack of formation of MoS2, which is consistent with the Raman spectra discussed earlier. Additionally, we found some new XPS bands in the range 160–180 eV that may be due to the S-related defects. It is expected from the CVD-grown samples that the additional bands from the 2p3/2 may be due to the S or Mo complexes. Due to low substrate temperature, the formation of polysulfides and ionized S (due to the weak bonding with Mo) is quite expected. The higher binding energy of S-2p peaks may correspond to the polysulfide or thiomolybdate (Spevack and McIntyre, 1993) (which has a core structure of MoSx where x ≠ y < 1) species. There may also be a formation of oxysulphide species, where S-O bonding is present, and this would produce the intense XPS bands in the range 167–170 eV. We also observed some additional peaks in the range 170–180 eV, which may be due to the presence of some impurities.

Figures 4G–I show the core level O-1s XPS spectra for the as-grown MoS2 dots. The O-1s spectrum has been fitted by two peaks (components) for all the samples. The peak position and the intensity ratio are summarized in Table 4. The peaks P1 and P2 are associated with the physisorbed oxygen and SiO2, respectively. We see that the ratio of the intensities of peaks P1/P2 for MS-560 is 0.11 and MS-510 is 0.17, while for MS-310 it is 6.44. This clearly reveals the presence of a higher content of physisorbed oxygen in the case of MS-310. This is consistent with the Mo (3d) XPS spectra, where we observed the presence of a higher integrated intensity of the Mo species associated with the MoO3 (see Figure 4, peaks at 235.7 and 237 eV). This is also consistent with the Raman analysis that showed the presence of the characteristic peak of MoO3 (Figure 2A).

Photoluminescence Studies

The as-grown MoS2 dots show strong visible PL at room temperature. Figure 5A shows the PL spectra of the MoS2 dots grown at different substrate temperatures (MS-560, MS-540, and MS-510) along with that of the commercial MoS2 flakes (MS). We observed a clear blue shift in the PL peak positions for the MS-560, MS-540, and MS-510 with respect to that of bulk MoS2. The blue shift is maximum for the MS-560 sample, which is due to the direct band gap transition in the monolayer MoS2, consistent with the previous reports. (Splendiani et al., 2010; Nan et al., 2014). Furthermore, the intensity of PL in MoS2 dots is about one order of a magnitude higher than that of the bulk MoS2 flakes. The strongly enhanced PL is direct evidence for the direct bandgap nature of the MoS2 dots, as compared to the indirect gap of the bulk MoS2. In order to understand the origin of the PL band, we have deconvoluted each spectrum of the MoS2 dot sample by using four Gaussian peaks (Figures 5B–D). The summary of the peak positions in each sample is listed in Table 5. Two characteristic PL peaks, P1 and P2, centered at ~621–631 nm and ~660–667 nm are ascribed to B exciton and A exciton, respectively. (Splendiani et al., 2010). An exciton peak is derived from the direct bandgap of MoS2, and the B exciton peak arises from the direct gap transition between the minima of the conduction band and the lower-level valence band maxima that is created by strong valence-band spin-orbit splitting at the K point (Splendiani et al., 2010). We also

### Table 3: Summary of the core level XPS peaks of Mo-3d and S-2p bands deconvoluted with Gaussian line shapes.

| Samples | Mo-3d | S-2p | Error bar (eV) |
|---------|-------|------|----------------|
| P1 (eV) | P2 (eV) | P3 (eV) | P4 (eV) | P5 (eV) | P1 (eV) | P2 (eV) | P3 (eV) | P4 (eV) | P5 (eV) | ±0.1 |
| MS-560  | 226.9 | 229.5 | 232.2 | 233.9 | 236.3 | 162.1 | 164.4 | 188.5 | 171.5 | 176.3 | ±0.1 |
| MS-510  | 226.9 | 229.1 | 232.2 | 234.4 | 236.5 | 162.2 | 164.4 | 186.2 | 173.1 | 176.9 | ±0.1 |
| MS-310  | –     | 230.5 | 233.0 | 235.7 | 237.1 | 162.4 | 164.8 | 188.0 | 171.5 | 177.2 | ±0.1 |

### Table 4: Summary of the XPS peak positions and their integrated intensity ratios for O-1s spectra of different samples.

| Sample | P1 (eV) | P2 (eV) | Integrated intensity ratio | Error bar (eV) |
|--------|--------|--------|---------------------------|----------------|
| MS-560 | 531.0  | 532.9  | 0.11                      | ±0.1           |
| MS-510 | 530.9  | 532.9  | 0.17                      | ±0.1           |
| MS-310 | 531.0  | 533.0  | 6.44                      | ±0.1           |
observed the presence of the $A^{-}$ exciton ($P_3$) peak, which is understood as arising from the trions. The trion emission is caused by light n-type doping from the substrate or from charged impurities in the CVD-grown MoS$_2$ dots. (Tongay et al., 2013; Dhakal et al., 2014). We also observed the presence of peak $P_4$ in all the samples at $\sim 696–700$ nm, and this peak is attributed to the radiative recombination of bound excitons. (Tongay et al., 2013). This defect peak may be due to the presence of S vacancies in the samples. Note that the relative intensity of $P_4$ is higher in the case of MS-510, than the other two samples. This is because of the lower temperature growth. All four peaks have been observed only in MS-560, MS-540, and MS-510. On the other hand, MS-310 did not show any PL, consistent with the lack of formation of MoS$_2$ as evidenced from the Raman and XPS spectra discussed earlier. We notice a small change in peak positions with the growth temperatures (see Table 5), which could be due to the non-uniformity in layer numbers for different samples.

**Figures 6A,B** show the position-dependent PL spectra of the MS-560 sample and the corresponding optical micrograph image showing the scanned area for each spectrum marked with a serial number. We observed that there is a no substantial shift in the PL peak position for different locations of the same sample, indicating fairly uniform growth of the MoS$_2$ dots. A marginal shift in the peak position at location 3 may be due to a small variation in the thickness of the MoS$_2$ dots. Previous reports suggest that the $P_2$ peak is blue-shifted, and the PL intensity is increased when decreasing the thickness from multilayer to monolayer MoS$_2$ films. (Mohapatra et al., 2016). Note that at positions 1 and 2, the integrated PL intensity ratio of the $P_2/P_1$ peak is much higher than that at the positions 3 and 4. We also observe a slight broadening of the PL peak in positions 3 and 4, and this may be due to the high trion emission, as the integrated intensity ratio of the $P_2/P_1$ peak is lowest in region 4. The $P_2$ peak decreases in intensity due to the enhanced nonradiative decay

**TABLE 5** | Summary of the PL peaks deconvoluted from the Gaussian line shape of the measured spectra for the different samples.

| Sample | $P_1$ (nm) | $P_2$ (nm) | $P_3$ (nm) | $P_4$ (nm) | Error bar (nm) |
|--------|------------|------------|------------|------------|----------------|
| MS-560 | 620.9      | 668.1      | 678.1      | 695.8      | ±1             |
| MS-540 | 630.7      | 666.7      | 683.2      | 699.8      | ±1             |
| MS-510 | 625.0      | 660.1      | 677.9      | 696.4      | ±1             |
| Identity | B exciton | A exciton | $A^{-}$ exciton/ Trion | Bound exciton |
when MoS$_2$ is doped with electrons. Since the growth is carried out at a relatively low temperature, the presence of structural defects and strain cannot be ignored. MS-540 and MS-510 samples show PL similar to that of MS-560 (see Supplementary Figures S3, S4). This visible PL at room temperature is a direct consequence of the direct gap nature of bilayer MoS$_2$. Thus, our results demonstrate that the CVD growth of an array of monolayer MoS$_2$ dots is possible at a temperature as low as 560°C. Note that monolayer CVD growth of MoS$_2$ is typically achieved at $\sim$1000°C, though the recent report shows that it can be grown at 680°C. However, the present report shows that monolayer CVD growth is achieved at 560°C.

**Growth Mechanism**

The growth mechanism of MoS$_2$ nanodots on the SiO$_2$ substrate is shown in Figure 7. Here, MoO$_3$ and sulfur are the precursors used and are placed at a particular distance away from the substrate as mentioned earlier. As the temperature of the single-zone furnace is ramped up to 800°C, first, the MoO$_3$ vapor is partially transferred by the Ar carrier gas and the MoO$_3$ molecules get attached onto the substrates. Next, as the sulfur vaporization takes place, the S atoms appear later near the substrates and react with the MoO$_3$ atoms adhered onto the substrate, resulting in the formation of MoS$_2$ ($2\text{MoO}_3 + 7\text{S}_2 \rightarrow \text{MoS}_2 + 3\text{SO}_2$). However, as the substrates are placed at the region of lower temperatures (510–560°C), due to the short diffusion length on the surface, the MoS$_2$ nanodots grown on the SiO$_2$ substrates are of non-uniform thickness, with a few tens of layers at the center. (Mun et al., 2016c). Additionally, the high flow rate of the Ar carrier gas throughout the experiment has also resulted in the formation of a uniform array of the MoS$_2$ nanodots.

**CONCLUSION**

Growth of highly luminescent bilayer and few-layer MoS$_2$ dots with controlled size and shape by a low-temperature (510–560°C) CVD technique was reported. The intense visible PL at room temperature with free excitonic emission indicated the direct gap nature of the crystalline MoS$_2$ dots array. Sulfur-induced surface defects are found and detected from the micro-Raman and XPS studies and correlated with the bound excitonic emission in the PL spectrum. Position-dependent Raman and PL profiles showed
uniformity in the MoS₂ layer for the growth at 540 and 560°C, while some nonuniformity in the MoS₂ layer was apparent for growth at 510°C. We believe the low-temperature growth of the luminescence MoS₂ dots array will stimulate further studies on growth on flexible substrates and its diverse range of applications in optoelectronics, bioimaging, and related areas.

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/Supplementary Material; further inquiries can be directed to the corresponding authors.

AUTHOR CONTRIBUTIONS

The work was conceived by all three authors. RKB and LPLM primarily conducted the experiments and analysis, and PKG interpreted the results. The manuscript was written and reviewed by all three authors.

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