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A Review of Physical Vapor Deposition of Two Dimensional Materials

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Abstract:

As the semiconductor industry demands higher chip integration, Moore's Law is increasingly constrained by physical limits of chip miniaturization and quantum tunneling effects. Traditional semiconductor materials can no longer meet the performance requirements of next-generation electronic devices. Two-dimensional materials, with their atomic-level thickness and exceptional physicochemical properties, have emerged as a critical breakthrough for overcoming these challenges. Physical vapor deposition (PVD), a core technology for synthesizing two-dimensional materials, offers advantages such as controllable deposition processes, high-fidelity thin film purity, and excellent compatibility with existing semiconductor processes, making it essential for large-scale production. This paper reviews common PVD methods, including thermal evaporation, electron beam evaporation, and sputtering (DC sputtering, RF sputtering, and magnetron sputtering), analyzes their pros and cons, and discusses innovations like contactless heating and pulse techniques. It also explores verified applications of PVD in cutting-edge research on two-dimensional materials such as graphene and TMDs. This review provides valuable references for optimizing PVD processes and improving equipment in two-dimensional material synthesis, while offering insights for future applications in semiconductor devices and optoelectronic detection.

Keywords: 2D materials; physical vapor deposition; evaporation; graphene; TMDs

1. Introduction

Since the emergence of Moore's Law in the 20th century, it has become the golden rule of the semiconductor industry, guiding the direction for every practitioner in this field. However, with technological advancements, the shrinking chip size and physical limitations have threatened this traditional guiding principle. The emergence of two-dimensional materials has become the most promising solution to address this dilemma. Materials ranging from graphene to transition metal dichalcogenides have demon-

strated practical application potential and significant development value. These two-dimensional materials not only possess ultra-thin thicknesses that meet current chip miniaturization requirements but also exhibit outstanding physicochemical properties: Tunable band gaps, unique optoelectronic characteristics, and more. This makes their application in semiconductor devices with smaller manufacturing processes feasible. In the research of two-dimensional materials, physical vapor deposition (PVD) stands as one of the most critical synthesis methods. This paper briefly introduces various PVD techniques and their applications in synthesizing verified two-dimensional materials, aiming to provide references for process optimization and equipment improvement in material synthesis. It also offers insights for future applications of two-dimensional materials in semiconductor devices, optoelectronic detection, and related fields.

2. Physical Vapor Deposition (PVD)

Physical Vapor Deposition (PVD) technology is a versatile method that utilizes mechanical, electromechanical, or thermodynamic processes to deposit materials onto substrates from their source [1]. Typically, this involves heating or bombarding a target material to vaporize it, which then adheres to the substrate surface. The heating methods, substrate properties, and surrounding environment significantly influence the film's characteristics. The two most common PVD techniques are evaporation and sputtering.

2.1 Evaporator Deposition

2.1.1 Thermal Evaporation

Thermal evaporation typically employs resistance heating elements to heat target materials under vacuum conditions. When the target reaches its boiling point, it vaporizes and deposits on the substrate surface. This process features relatively simple equipment and convenient operation, making it effective for low-melting-point targets. However, it proves challenging for high-melting-point materials. Additionally, the contact between the crucible and target during heating may cause contamination. Controlling the evaporation rate also presents significant difficulties.

Ioannis A. [1] et al. introduced a novel physical vapor deposition (PVD) system for thin film fabrication (Figure 1). This innovative device utilizes high-frequency current and zero-voltage switching (ZVS) heaters to heat sublimation targets without physical contact, effectively preventing material contamination caused by crucible sublimation. The ZVS circuit design also reduces energy consumption, achieving lower operational costs. However, it should be noted that while this system does not require low-melting-point materials like conventional thermal evaporation equipment, the target materials must possess excellent electrical conductivity.

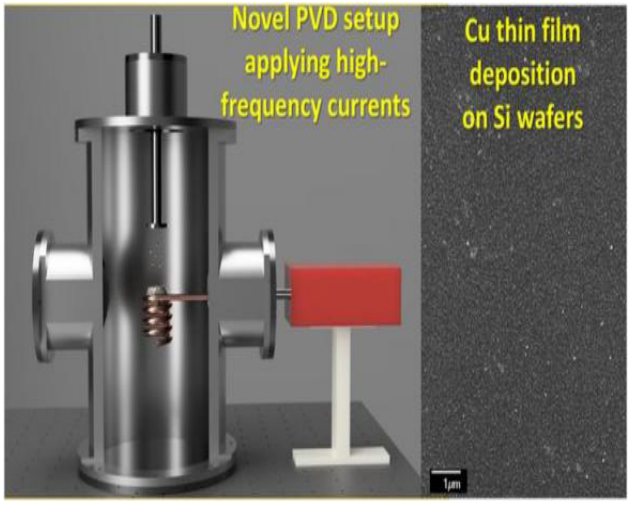

Fig.1 High-frequency current ZVS vapor deposition device [1]

2.1.2 Electron Beam Evaporation

Electron beam evaporation utilizes a focused electron beam directed at the target material to vaporize it. By employing high-energy electrons, this method achieves superior evaporation control compared to conventional thermal evaporation, ensuring high-quality and uniform thin film deposition. Notably, it can process both low-melting-point materials and high-melting-point targets. However, the technique faces challenges, including high equipment costs and the need for precise electron beam energy control to prevent material damage [2].

Jedrzejewski, R. [3] and colleagues pioneered the use of pulsed electron beam deposition (PED) to fabricate polymer-carbon composite thin films (Figure 2). Their PED-coated structures with composite architectures demonstrated the technique's capability to completely remove complex materials—including nanotubes—from targets and deposit them onto substrates. This breakthrough deepened understanding of electron beam evaporation deposition and composite material deposition mechanisms, paving the way for novel applications of advanced sensor composite films.

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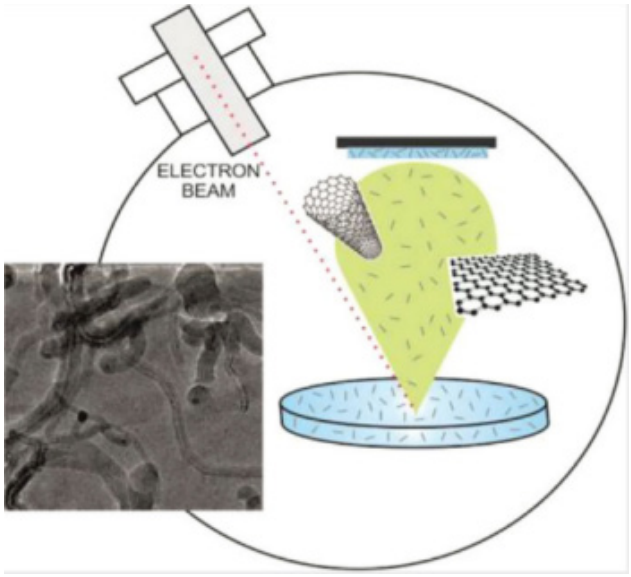

Fig.2 Pulsed electron beam deposition (PED) fabricates polymer-carbon composite thin films [3]

2.2 Sputtering

Sputtering deposition is a widely used thin-film deposition technique in chip manufacturing. Typically employed in semiconductor production to deposit various metals, dielectrics, and thin films on wafers, this process involves introducing inert gas into a chamber. The gas is ionized into positive ions and free electrons, which collide with the target material surface under an electric field. These high-energy particles eject target atoms, depositing them onto the wafer substrate to form desired thin films. Sputtering can be categorized into DC sputtering, radio frequency sputtering, magnetron sputtering, and reactive sputtering. This method effectively addresses issues like uniformity and quality control in evaporation processes, and is classified as part of physical vapor deposition (PVD) technology.

The most common method for thin film deposition through sputtering involves using a magnetron source, where positive ions in the magnetron-enhanced glow discharge plasma bombard the target. Since sputtering is a purely physical process, it must temporarily introduce reactive gases into the plasma to deposit compound layers by reactive sputtering. The magnetic assembly forms a magnetic bottle. The magnetic field causes the secondary electrons emitted from the target material to move in a spiral motion on the surface of the target material, increasing the probability of them colliding with the working gas and undergoing ionization [4].

Chen, XP. [5] et al. utilized a radio frequency (RF) magnetron sputtering system to deposit MoS_2 thin films on fluorine-doped tin oxide (FTO) glass substrates under argon gas flow. They compared MoS_2 films prepared at sputtering pressures of 4Pa, 2Pa, 0.8Pa and 0.5Pa. XRD, SEM, and AFM analyses revealed that the MoS_2 film deposited at 0.8Pa exhibited the smallest size and most uniform lamellar structure (Figure 3). This phenomenon may be attributed to the faster deposition rate of MoS_2 at 0.8Pa, which facilitates more nucleation and results in smaller lamellar particles.

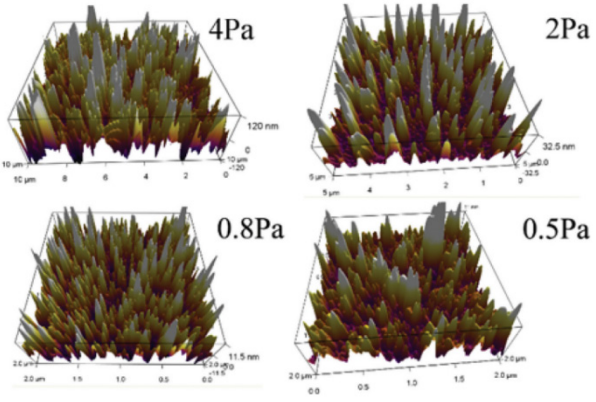

Fig.3 AFM images of MoS_2 films prepared under different sputtering pressures [5]

3. Research on PVD-based Two-dimensional Materials

As of this writing, numerous two-dimensional materials have been confirmed to exist at room temperature. Their exceptional thermal and chemical stability makes them ideal as solid lubricants. Notable examples include graphite, hexagonal boron nitride (hBN), and monolayer graphene (MoS_2), which have been extensively studied. Additionally, various oxide monolayers warrant further investigation.

3.1 Graphene

Yuan, BN. [6] et al. reported a simple low-temperature PVD technique for depositing graphene-on-carbon (g-CN) films on biomass substrates. While PVD is a straightforward method for preparing graphitic carbon nitrides, existing vapor deposition techniques require temperatures exceeding 500℃, severely limiting their application on biomass substrates. Photoelectrochemical experiments

demonstrated that the g-CN films prepared by this method exhibit identical light-responsive properties to conventional g-CN films. Moreover, life cycle assessments revealed that biomass substrates have significantly lower environmental impacts compared to traditional ITO glass substrates. This study paves the way for the widespread application of biomass-based materials in semiconductor optoelectronics.

Garlow, JA. [7] et al. successfully grew large-area turbine-layer graphene on heterogeneous epitaxial Ni (111) films by precisely controlling the deposition temperature, beam flux, and total deposition carbon amount. By comparing the graphene regions under different deposition temperatures and spatially identifying the large-area turbine-layer graphene on the nickel film using PVD, they provided a unique opportunity to explore the basic characteristics and feasibility of turbine-layer graphene for future applications such as data storage and transmission technologies.

Vijayaraghavan, RK. [8] et al. demonstrated the growth of single-layer and multi-layer graphene on copper foil using bipolar pulsed DC magnetron sputtering in a pure argon atmosphere, proposing a diffusion-controlled graphene growth mechanism (Figure 4). Their findings revealed that platelet adhesion, activation, and aggregation on graphene-coated surfaces were significantly inhibited compared to bare glass, indicating that the deposited graphene films exhibit anti-coagulant activity.

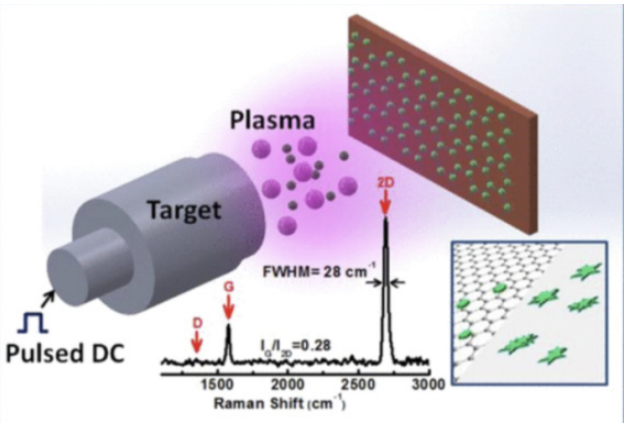

Fig.4 The growth of single-layer and multi-layer graphene on copper foil using bipolar pulsed DC magnetron sputtering in a pure argon atmosphere [8]

3.2 Two-dimensional Transition Metal Chalcogenides (TMDs)

Rahman, MS. [9] and colleagues investigated the deposition of various metals on MoS_2 using two physical vapor deposition (PVD) techniques—electron beam evaporation and magnetron sputtering—while examining the impact of metallization on MoS_2's vibrational and electronic properties. Their Raman spectroscopy analysis revealed that sputtering-induced degradation in single-layer MoS_2 was more severe than that caused by electron beam evaporation. Furthermore, comparative studies showed that evaporated Sn/Au contacts exhibited superior performance metrics in field-effect transistors (FETs) compared to sputtered counterparts, demonstrating lower contact resistance, enhanced mobility, and improved subthreshold slope. These findings provide valuable insights for developing transition metal dichalcogenide (TMD) devices.

Madoune, Y. [10] et al. highlighted the critical importance of optimizing fundamental growth conditions for producing high-quality materials. Through systematic investigation of key parameters including temperature, powder dosage, gas flow, growth duration, gas flow direction, and substrate positioning, they achieved remarkable growth of WS_2 films with 850 μm thickness by adjusting the source quantity to 0.5 g, gas flow rate to 120 Sccm, and growth time to 10 min at 1180℃. These findings provide valuable references for achieving high-quality WS2 film growth.

Liu, XY. [11] et al. synthesized high-quality two-dimensional WSe_2 materials through physical vapor deposition (PVD) and chemical vapor deposition (CVD) methods. Comparative electrical characterization of transistor devices demonstrated superior optoelectronic performance in PVD-synthesized materials, while the semiconductor conductivity exhibited opposite characteristics between the two processes. Photoemission spectroscopy (PL), Raman spectroscopy, and X-ray photoelectron spectroscopy (XPS) analyses revealed distinct conduction types: PVD-derived WSe_2 displays p-type semiconductor properties with enhanced electrical performance and improved electrode-material interface, whereas CVD-derived WSe_2 exhibits n-type semiconductor characteristics with inferior electrical performance. These differences in conduction types enable the fabrication of high-performance PN junctions and heterojunction photonic devices, paving the way for broader applications in optoelectronic control systems with promising development prospects.

Tianjun Dai [12] and colleagues developed a straightfor-

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ward PVD method using an electron beam evaporator to directly deposit $MoSe_2$ precursors. The molecular $MoSe_2$ compounds were transferred to the growth surface, enabling epitaxial growth on graphene (a van der Waals material) that enhances surface diffusion and facilitates large-scale heterostructure fabrication. TEM measurements revealed that $MoSe_2$ can achieve large-area continuous epitaxy on CVD-grown graphene via PVD. Unlike conventional $MoSe_2$ deposition methods that produce random orientations, the $MoSe_2$ film predominantly grows on graphene in the $MoSe_2$/graphene system, with AFM images showing 214 pm RMS roughness, demonstrating excellent uniformity. This breakthrough not only enables large-area vertical $MoSe_2$/graphene heterostructures but also suggests that controlled-oriented two-dimensional TMDCs van der Waals heteroepitaxy could introduce effective interlayer interactions, potentially boosting their photonic applications.

3.3 Other Materials

Xu, X. [13] et al. fabricated a series of two-dimensional SnS thin films with varying thicknesses via physical vapor deposition (PVD) and characterized them using Raman spectroscopy, X-ray diffraction (XRD), and X-ray photoelectron spectroscopy (XPS). The results demonstrated that the prepared films exhibited high crystallinity and low defect density. Based on these findings, they designed a SnS-based photoelectrochemical photodetector that not only achieved higher Iph (5.50 $\mu A/cm^2$) and Rph (16.34 A/W) photocurrents but also exhibited self-powered optical response at 0.6 V. The study highlighted the significant potential of two-dimensional SnS thin films in photodetector applications, owing to their superior performance characteristics, including high absorption coefficients, narrow band gaps, and excellent carrier mobility.

Sondors, R. [14] et al. optimized a simple catalyst-free physical vapor deposition (PVD) method by adjusting source material pressure and evaporation time, enabling reliable fabrication of independent nanoribbons with thicknesses below 15 nm. They identified the optimal synthesis conditions (temperature, time, and pressure) for producing ultra-thin Bi_2Se_3 nanoribbons with thicknesses ranging from 8 to 15 nm.

Huang, TY. [15] and colleagues developed a substrate-engineered PVD method to synthesize high-quality Te nanosheets, achieving a field-effect hole mobility of 1450 $cm^2/(V \cdot s)$. The high-mobility p-type Te nanosheets synthesized by this method enabled the fabrication of van der Waals (PVN) heterojunction photodiodes. Demonstrating that Te-MoS_2 synthesized via PVD exhibits photodetection capability with a current response rate of up to 630 A/W, the team found that the current response speed achieved using p-type Si-MoS_2 is one order of magnitude faster. Additionally, the Te-MoS_2 photodiodes demonstrated excellent gate tunability and unique optical properties. This innovative PVD synthesis method and the demonstrated Te-MoS_2 photodetector open new possibilities for developing ultra-lightweight and highly tunable optoelectronic devices.

Wang, YG. [16] et al. introduced a sealed space composed of quartz interlayers during the PVD growth of two-dimensional PbI_2. This novel spatially confined PVD method provides enhanced control over growth kinetics, enabling the production of highly uniform triangular PbI_2 flakes. By fine-tuning the sealed space height and substrate distance, controllable uniform PbI_2 growth allows fabrication of flakes with adjustable thickness and lateral dimensions (Figure 5). The grown two-dimensional PbI_2 flakes demonstrated outstanding performance in photodetectors, exhibiting 0.72 A/W high responsivity, a switching ratio of up to 90,013.5 ms fast response time, and a 20 ms decay time. The newly developed enhanced spatially confined PVD method not only achieves controllable preparation and high-yield synthesis of two-dimensional materials but also advances the in-depth development of optoelectronic applications.

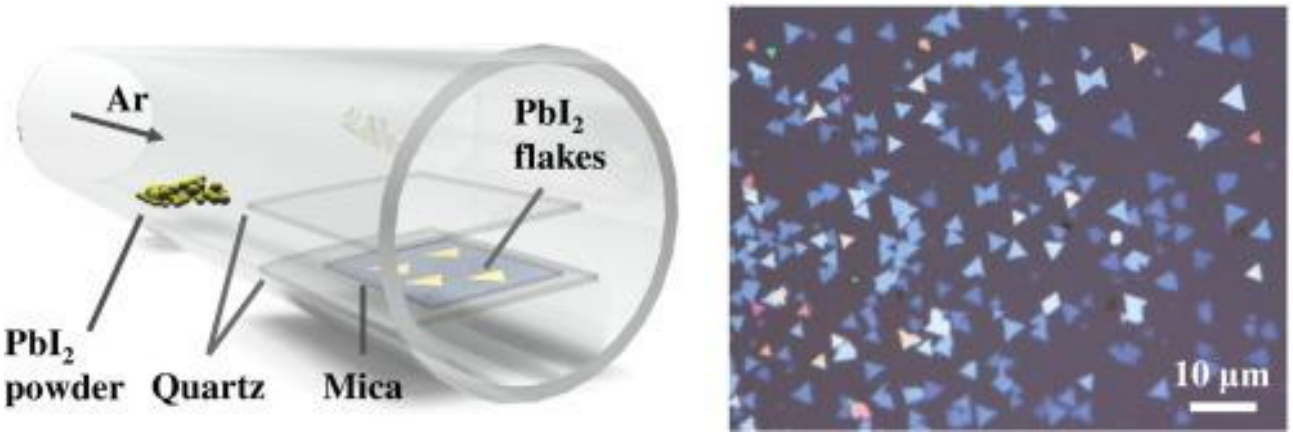

Fig.5 The synthesis of PbI_2 triangle flakes with high homogeneity [16]

4. Conclusion

Physical vapor deposition (PVD) technology, as a crucial method for synthesizing two-dimensional materials, has provided robust support for optimizing material properties and expanding applications through its diversified methodologies and continuous technological innovations. As two core PVD techniques, evaporation and sputtering each demonstrate unique advantages and limitations. Through ongoing innovations (such as contactless heating and pulse technology applications), these methods continuously address their inherent constraints while enhancing deposition efficiency and material purity. In terms of applications for two-dimensional materials, this paper introduces PVD implementations in graphene materials, two-dimensional transition metal sulfides, two-dimensional SnS films and Te films. For graphene materials, precise control of deposition temperature and beam flux parameters enabled large-area heteroepitaxial growth of turbostratified graphene on Ni (111) films via PVD technology. Furthermore, under a diffusion-controlled graphene growth mechanism, single-layer and multi-layer graphene with anti-coagulation activity were successfully fabricated on copper foil. In the field of two-dimensional transition metal sulfides, PVD technology has been applied to synthesize and metallize materials like MoS_2 and WS_2, with devices prepared by electron beam evaporation demonstrating superior contact resistance and mobility. For other two-dimensional materials, PVD-synthesized SnS films exhibit excellent nonlinear optical properties, and process optimization supports their applications in nonlinear photonics and magnetic transport. The synthesis of Te nanosheets via PVD has also paved the way for the development of p-type semiconductors in van der Waals heterostructures and photodiode advancements. The application of these PVD technologies in various materials provides relevant references for the preparation and application of subsequent 2D materials.

The future of PVD technology in 2D materials holds vast potential. On one hand, continuous process innovation is crucial to reduce equipment costs, enhance deposition rates and material compatibility, and overcome bottlenecks in large-scale production of high-quality 2D materials. On the other hand, research should focus on aligning PVD processes with 2D material properties to achieve synergistic optimization between material preparation and device integration, addressing performance requirements in target applications like semiconductors. As the technology matures, PVD will drive industrial applications of 2D materials across more high-tech sectors, injecting sustained momentum to break through performance limitations of traditional materials and propel technological innovation in related industries.

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