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学 位 の 種 類 博士(工学)

学 位 記 番 号 博第1220号

学位授与の日付 2021年9月1日

学位授与の条件 学位規則第4条第1項該当 課程博士

学位論文題目 Growth of molybdenum disulphide on semiconductor substrates

for heterojunction device applications

(半導体基板上への二硫化モリブデンの合成とヘテロ接合デバイス

への応用)

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論文内容の要旨

In this thesis work, I have emphasized on the growth of transition metal dichalcogenides (TMDCs) layers on Si and GaN semiconductor substrates by the chemical vapour deposition (CVD) technique. The heterostructures of molybdenum disulphide (MoS₂) with Si and GaN were analysed by the device fabrication and their suitability for optoelectronics were explored.

<u>Chapter 1</u> describes the background of TMDCs materials and their unique properties. Various synthesis methods of TMDCs layer and their characterizations process to identify the layer structures, stability issues and current trend of device applications are discussed. Then, the challenges in the synthesis of mono and few layers of TMDCs and their continuous films were introduced.

<u>Chapter 2</u> deals with the synthesis processes for the growth of MoS₂ layers and crystals. The CVD techniques used in these studies for the growth of MoS₂ layers were presented and discussed in detail. The physical and chemical characterisation methods of MoS₂ based 2-dimensional materials were introduced in this chapter. These characterisation techniques are used in ascertaining the chemical composition, structure and so on for the synthesised MoS₂.

Chapter 3 describes the fabrication of MoS₂ and p-type Si heterostructure using molybdenum oxide (MoO₃) as the precursor. In this synthesis process, MoO₃ layer was directly deposited on Si and sulphurization was performed in a CVD process. A heterojunction device fabricated for the deposited MoS₂ layers on the Si substrate showed a photovoltaic action and a photoresponsivity of 139 mA/W at 860 nm wavelength for a bias voltage of ·5V. Then, I discuss the effect of interface states in the MoS₂/Si heterojunction and the influence in photoresponsivity. The understanding of photocarriers behaviour in the fabricated MoS₂/Si heterojunction interface can be critical to develop high photoresponsive devices.

Chapter 4 deals with the feasibility of van der Waals (VdW) heteroepitaxy of MoS2 layers on a lattice matched GaN semiconductor. The uniform MoS₂ layers were directly grown on the n-type GaN wafer by sulphurisation process of MoO3 thin layer using the CVD process employed in the previous chapter. Homogenous growth of the few-layers MoS₂ forming a continuous film was achieved, signifying the suitability of GaN semiconductor substrate. The fabricated MoS₂/GaN vertical heterojunction showed the excellent rectifying diode characteristics with a photovoltaic action. Then, the band structure for the MoS₂/GaN heterojunction was investigated, confirming a so-called type II-based heterojunction. In addition, analyses of the growth of MoS₂ crystals on the lattice matched Ga-polar GaN wafer using ammonium tetrathiomolybdate (ATM) as a new precursor in a CVD process, instead of using the molybdenum oxide-based precursors, are demonstrated. Unidirectional triangular MoS₂ crystals and continuous film were obtained on the free-standing Ga-polar GaN substrate in this CVD process. Then, a heterojunction device was fabricated for the synthesised MoS₂ layers on GaN substrate, obtaining better diode rectification behaviour compared to device fabricated using the CVD with MoO3 as a precursor.

<u>Chapter 5</u> summarizes the whole thesis and future prospects.

論文審査結果の要旨

Transition metal dichalcogenides (TMDCs) are one of the hottest materials in nanoscience and nanotechnology, and a variety of applications has been proposed. In this thesis, the growth of MoS₂ on Si and GaN semiconductor substrates by the chemical vapor deposition (CVD) technique is dealt with, together with the vertical heterojunction device application for optoelectronics.

Chapter 1 includes the introduction to the fundamentals and applications of TMDCs of monoand few-layers and their continuous films, as well as the motivation and the purpose of the thesis.

Chapter 2 deals with the synthesis and characterization methods used in this thesis in detail for MoS₂ which is one of the typical TMDC materials.

Chapter 3 describes the synthesis of MoS₂ layers on a p-type Si substrate using molybdenum oxide (MoO₃) as the precursor to fabricated the heterostructure. In this CVD synthesis process, MoO₃ was directly deposited onto Si and subsequent sulfidation was performed. A heterojunction device fabricated for the deposited MoS₂ layers on the Si substrate showed a photovoltaic action and a photoresponsivity of 139 mA/W at 860 nm in wavelength for a bias voltage of -5V. In addition, the effect of interface states in the MoS₂/Si heterojunction and the influence in photoresponsivity are discussed. The understanding of photocarriers behavior in the fabricated MoS₂/Si heterojunction interface can be critical to develop high photoresponsive devices.

Chapter 4 deals with the feasibility of van der Waals (VdW) heteroepitaxy of MoS₂ layers on a lattice matched GaN semiconductor. The uniform MoS₂ layers were directly grown on the n-type GaN wafer by the sulfidation of MoO₃ thin layer using the CVD process employed in the previous chapter. Homogenous growth of the few-layers MoS₂ forming a continuous film was achieved, signifying the suitability of GaN as a semiconductor substrate. The fabricated MoS₂/GaN vertical heterojunction showed the excellent rectifying diode characteristics with a photovoltaic action. The investigation of the band structure for the MoS₂/GaN heterojunction confirms a so-called type II band alignment in the fabricated heterojunction device. For a comparison, the growth of MoS₂ crystals on the lattice matched Ga-polar GaN wafer using ammonium tetrathiomolybdate (ATM) as a new precursor in a CVD process, instead of using the MoO₃ precursor, is also attempted. Unidirectional triangular MoS₂ crystals and continuous film were obtained on the free-standing Ga-polar GaN substrate in this CVD process. A heterojunction device fabricated for the synthesized MoS₂ layers on the GaN substrate showed a better diode rectification behavior compared to the heterojunction device fabricated using the MoS₂ starting from MoO₃ precursor.

Chapter 5 summarizes the whole thesis and future prospects.

Thus, this thesis demonstrates the new findings on the synthesis of MoS₂ on Si and GaN semiconductor substrates and on their application to the photoresponsive devices. This is enough worth for PhD thesis.