Semiconducting Metal Oxide Thin-Film Transistors

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Wenfeng Zhang and Xiaokun Wen

Preface

Semiconducting metal oxide thin-film transistors are promising candidates for functional electronic devices. They have attracted considerable attention owing to their superior electrical performance, high transparency, excellent stability and uniformity. Semiconducting metal oxides usually exhibit higher carrier mobilities than those of amorphous Si and most organic transistors, and better device uniformity than that of polycrystalline Si transistors.

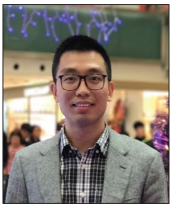
In this book, we will first introduce the concept and basic working mechanism of semiconducting metal oxide thin-film transistors. We then focus on a series of metal oxide thin films with desirable electrical and optical properties for various device applications. The typical devices such as logic circuits, LED drivers, photodetectors, tactile imaging, transistor memories, artificial synaptic devices and transparent transistors will be discussed.

I would like to express my gratitude to all the fellow authors who have contributed in this book. I also want to acknowledge Robert Trevelyan and Caroline Mitchell at IOP Publishing, for all the help during the book editorial process, and for the excellent experience of working with them. I want to thank all the readers for their interest in our book. I hope that our book can be useful as a reference guide for researchers and students who work in the field of metal oxides and metal oxide-based transistors.

Ye Zhou

Editor biography

Ye Zhou



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Chapter 1

Introduction to semiconducting metal oxides

Shi-Rui Zhang and Ye Zhou

Semiconducting metal oxide (SMO) materials are promising candidates for various electronic devices. They have attracted considerable attention owing to their superior electrical performance, high transparency, excellent stability and uniformity. In this chapter, we mainly introduce some basic ideas of SMOs.

1.1 Background

Oxide, containing at least one oxygen and one other element, is one of the most abundant compounds on Earth, existing in gas, liquid and solid all over the place. A very common example of liquid oxides in room temperature is hydrogen oxide (H_2O) which we call water, used as a universal solvent and necessary for life on our blue planet. Carbon dioxide (CO_2) is a kind of gas oxide familiar to people, known as an ingredient of photosynthesis and one of the products of respiration. Apart from water and gas oxide, solid oxides also play an important role in our daily life. For instance, silicon oxide (SiO_2), also called silica, has widespread applications in daily necessities, architectures and technologies. The most traditional type of colorless glass is quartz, in which silicon oxide is the single constituent in amorphous state. With excellent optical properties and chemical stability, quartz is not only utilized to manufacture optical fibers used in cable television and communication systems but also applied to the processing of diverse semiconductor devices [1]. Since the advent of the first MOSFET in the late 1950s, solid oxides have been proved to be indispensable in the modern semiconductor industry [2, 3]. The aforementioned silicon oxide acts as an insulating layer in the metal-oxide-semiconductor (MOS) structure, the core of MOSFET. MOSFET is a kind of device with three terminals: source, drain and gate. The MOS structure inside the device can be regarded as a planar capacitor where the semiconductor is basically p-type or n-type silicon and the metal layer could be conductive polycrystalline silicon besides metals. At the beginning, MOS structure can be fabricated by growing a layer of silicon oxide through thermal deposition on the silicon wafer and then depositing the metal layer.

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When talking about the function of oxide layer in MOS structure and even in the whole device, we could simply say that it performs a dielectric property, preventing electrons or holes from tunneling from semiconductor to metal gate, contributing to the formation of channel near the interface between oxide and semiconductor.

Alhough oxides were usually thought to be insulators, some metal oxides such as simple binary oxide including ZnO, In_2O_3, SnO_2, etc [4], and complicated oxide Zn–Sn–O, In–Sn–O [5, 6] actually have properties of semiconductors. Before going deep into talking about SMOs, a widely used device should be introduced. As the derivative of MOSFET, thin-film transistors (TFTs) have wide application in electronics. TFT works based on the same field effect mechanism as MOSFET. In general, TFT is fabricated by deposited semiconductor materials, dielectric materials and conducting materials in the form of thin film on a certain substrate [7–9]. The primary application of TFT is in display products such as liquid crystal display (LCD) and organic light-emitting diode (OLED) which are based on active matrix (AM) [10, 11]. TFT is widely used in the manufacture of active matrix liquid crystal display (AMLCD), occupying the largest segment of output of the panel display industry. In the meantime, active matrix organic light-emitting diode (AMOLED) is in rapid development. Generally speaking, TFT used in AMLCD and AMOLED can be made by using a very wide variety of semiconducting materials, but commonly used material is silicon including single crystalline silicon, microcrystalline silicon, low temperature polysilicon (LTPS) and amorphous silicon. Facing the advent of next generation display technologies, there are still some disadvantages existing in current silicon based TFT for display. To begin with, amorphous silicon based TFTs have relatively inadequate mobility, lower than 1 $cm^2\ V^{-1} \cdot s^{-1}$, limiting the operating speed of TFT devices as higher mobility could lead to high current which makes capacitive loads quickly charged or discharged [12, 13]. Besides, though LTPS based TFTs possess mobility about 100 $cm^2\ V^{-1} \cdot s^{-1}$, the existence of lots of grain boundaries leads to the difference of electrical characteristics between TFT units [14, 15]. What's more, silicon based TFTs are opaque to visible light and temperature of deposition is generally high, not applicable to the trend of transparent display and flexible panels. By using transparent electrodes such as indium tin oxide (ITO) and transparent semiconductors we can make TFTs totally transparent. In this condition, semiconducting metal oxide becomes an ideal candidate due to its sufficient mobility and characteristics of transparency under visible light, attracting great attention from researchers. In 2003, several groups adopted ZnO as n-channel to fabricate highly transparent TFTs [16–18]. It's worth noting that these ZnO-based TFTs could reach mobility larger than 1 $cm^2\ V^{-1} \cdot s^{-1}$, symbolizing the beginning of application of SMOs as the semiconducting channel layer in TFTs.

1.2 Fundamentals of semiconducting metal oxide

As mentioned above, oxide contains at least one oxygen and one other element, existing in solid, liquid and gas phase. Unlike molecular oxides, SMO generally has the form of solid and belongs to the family of non-stoichiometric compounds, which means proportions of elemental composition cannot be exactly represented by a ratio of small natural numbers. As a kind of ionic solid, SMO has strong ionic bonds connecting positive metallic ions and negative oxygen ions. As for SMO, inside the materials, the 's' electronic shell is filled inherently while the 'd' shell is incompletely filled, owing to which, SMO possesses superior thermal/chemical stability and unique properties such as tunable energy bands, high dielectric constants and so on [19]. And these unique properties endow SMO capabilities in different applications. SMO is a very large group of materials, so they differ from each other in crystalline structure, inherent defects, energy band level and other physical aspects. For example, in terms of crystalline structure, ZnO could exist in hexagonal wurtzite or cubic zincblende. However, under ambient conditions, hexagonal wurtzite is more stable and common with the space group of $P6_3mc$ [20]. The lattice constants of ZnO in wurtzite form are $a = 3.25$ Å and $c = 5.2$ Å, where the ratio of c/a is 1.633, closed to ideal value. In contrast, In_2O_3 crystallizes in the form of cubic with lattice constant of 10.08 Å while SnO_2 belongs to rutile tetragonal and lattice constants are $a = 4.737$ Å, $c = 3.185$ Å [4]. These various crystal structures lead to different unique properties of SMO.

For oxides, several kinds of defects exist widely inside materials, including point defects, line defects, plane defects and volume defects. In fact, defects exist in all crystalline solids which are not perfect though they exhibit periodic crystal structures [21, 22]. Point defects are also called zero-dimensional defects and can be mainly divided into three kinds, including vacancy defects, interstitial defects and Frenkel pairs when there is no heteroatom [23, 24]. Vacancy defects refer to vacant sites that should be occupied by certain atoms in a totally perfect crystal. And because of the stability of crystal structure around the vacancy, the neighboring atoms of the vacancies will not collapse easily, guaranteeing the existence of these vacancies. What's more, once a certain atom moves into the vacant site, the vacancy would move in the opposite direction and occupy the site where the atom used to be located. In contrast, when atoms appear in a region where there are commonly no atoms existing, interstitial defects show up with generally high energy configuration. The last kind of defect seems to be a combination of vacancy defects and interstitial defects, which means an atom occupies an interstitial site and leaves a vacancy, called Frenkel defects. As mentioned above, SMO belongs to ionic solids, owing to which, ionic bonds are more easily broken than covalent bonds so SMO usually has more intrinsic defects compared to elemental semiconductor materials [25]. For SMO, there are six classic point defects such a site of metallic atom (M) replaced with oxygen atom (O), a site of O replaced with M, no atom on a site of M, no atom on a site of O, M or O existing in an interstitial region [19]. On account of the existence of intrinsic defects like oxygen vacancies and metallic interstitial, undoped SMO generally exhibits n-type semiconducting properties. But for certain SMOs, the origin of n-type semiconducting is still controversial. For instance, there is another explanation for n-type conductivity of ZnO instead of inherent defects in the absence of intentional doping [26]. By using first-principle calculation and experiment, hydrogen is identified and characterized in ZnO bulk, being thought to be responsible plausibly as its presence is inevitable in most growth and annealing processes [27–30].

For solid-state physics, band theory is a significant concept, describing the range of energy levels that electrons could stay at (bands) and could not stay at (band gap). Pauli exclusion principle finally leads to the formation of valence band and conduction band [31, 32]. Many physical properties of solids including resistance, absorption and so on can be well explained by band theory. In short, band gap means a range of energy where no electronic states exist and band gap energy of a semiconductor is the minimum energy that the electrons required to transfer from valence band into conduction band. According to band theory, electrons cannot move freely inside the solid if the valence band is totally full and the conduction band is completely empty. Once electrons transfer from valence band to conduction band, they will be able to move freely, which means the formation of current. External heat or a photon with energy higher than band gap energy could excite electrons so that they can leave the valence band and transit into the conduction band, leaving orbital holes in the valence band. For most SMOs, the band gap is generally wide (>3 eV). For example, the band gap of ZnO, In_2O_3, SnO_2 is about 3.4 eV, 3.5 eV, 3.6 eV [4]. Owing to the wide-band gap, SMO materials are able to endure large electric field and have high breakdown voltages, which means that SMO-based electronic devices could be operated under high temperature and power. What's more, the wide band gap also endows SMOs with high optical transparency in the visible region of the electromagnetic spectrum, which is a desired characteristic in transparent electronics.

In addition to band theory, impurity level is another vital parameter of semiconductors. Impurity level has significant impact on carrier concentration, microstructure and related aspects of a semiconductor. Generally, through doping, the intentional introduction of extra compositions into an intrinsic semiconductor alters the impurity level, resulting in the variation of electrical and optical properties. From the perspective of band theory, doping introduces extra energy levels within the band gap and the introduced energy levels will be closed to valence band or conduction band when species of dopant are different. Specifically, electron donor or n-type doping brings in energy levels near the conduction band while electron acceptor or p-type doping brings in ones near the valence band. As for ZnO, n-type doping can be achieved by replacing with Al, Ga, In and other group-III elements or by substituting group-VII elements for O while p-type doping is commonly thought to be difficult [4, 33, 34]. From point of view of carrier concentration, regardless of n-type or p-type doping, doped semiconductors would have higher carrier concentration compared to their intrinsic state because both electrons in n-type semiconductors and holes in p-type semiconductors contribute to conductivity significantly. Some highly doped semiconductors even possess conductivity comparable to metals. The reason impurity level of semiconductor is importants is that it has a direct impact on the carrier mobility, which is another crucial parameter of semiconductors when they are applied to electronic devices. Carrier mobility is related to two kinds of scattering inside material: lattice scattering (phonon scattering) and ionized impurity scattering [35]. In terms of lattice scattering, when temperature is higher than absolute zero, atoms inside semiconductor will vibrate and create acoustic waves (phonons), which break the ideal periodic potential. As a consequence, vibrating atoms interact with moving carriers by scattering them, causing the reduction of mobility. In other words, higher temperature results in smaller mobility and vice versa. Regarding ionized impurity scattering, electron donors or acceptors are commonly ionized when being doped into semiconductor. Coulombic interaction will be produced when moving electrons or holes approach these doped impurities, leading to scattering of carriers. Under constant temperature, increasing impurity level normally lowers the carrier mobility.

1.3 Application of semiconducting metal oxide

SMOs are widely used in many fields, especially in electronics. The electrode may be the simplest application of SMO. For example, indium tin oxide (ITO) is a highly doped n-type semiconductor with low resistivity and high carrier concentration. Besides, ITO also possesses typical wide band gap and high transmittance in the range of visible light. These unique electrical and optical properties make ITO an ideal candidate for a transparent conducting electrode [36]. For example, ITO can not only be drain, source and gate electrode in TFT, but can also be used as anode in OLED [6, 37, 38]. Furthermore, due to the transparent conductive property as well as ease of being deposited as thin film, ITO is used to make transparent conductive coating for display technologies and photovoltaics [39–41].

As mentioned above, TFT is one of the important applications of SMO. The introduction of SMO could break the limitation of traditional silicon based TFT. In 2003, Hoffman *et al* [17] investigated transparent thin-film transistor (TTFT) devices by using ZnO as n-channel. In this research, highly transparent n-type ITO served as drain, source and gate electrode at the same time with aluminum–titanium oxide (ATO) deposited as gate insulator. These TTFTs showed optical transmission of 75% in the visible light region of the electromagnetic spectrum and drain current ON/OFF of about 10^7. Most importantly, this research also demonstrated that these ZnO based devices' channel mobilities could be more than 1 $cm^2\ V^{-1} \cdot s^{-1}$ and reached up to 2.5 $cm^2\ V^{-1} \cdot s^{-1}$. Nomura *et al* [18] fabricated TFTs in single-crystalline highly doped oxide semiconductor, $InGaO_3(ZnO)_5$. The devices exhibited field-effect mobility of about 80 $cm^2\ V^{-1} \cdot s^{-1}$ due to less grain boundary and scattering in a single crystal as well as high doping density. In fact, in the modern display industry, amorphous In–Ga–Zn–O (a-IGZO)-based TFTs perform excellently due to high mobility and high uniformity, so a-IGZO is a kind of material being widely used in actual mass production [42–47].

The use of SMO-based TFT was naturally extended to the area of flash memory devices as they adopted TFT as fundamental structure. In 2017, Hota *et al* [48] constructed a transparent flash memory by combining amorphous oxide with high-κ oxide materials by the sol–gel method. As a kind of n-type semiconductor, ZnO is applied to form the channel. Ta_2O_5 functions as the charge trapping layer and tunneling dielectric. For low voltage operation, Al_2O_3 plays the role of blocking dielectric layer. ITO is the electrode. With program and erase, the maximum memory window of ~10.7 V is demonstrated. Flash memory performed well in durability and reliability tests, holding data for more than 10^4 s and having approximately 100 program/erase cycles. With the development of non-volatile memory devices, resistive random access memory (ReRAM) is thought to be one of the potential candidates for next-generation devices. So many materials exhibit resistive switching phenomenon which is required in ReRAM. In fact, resistive switching phenomenon was first found on Al_2O_3 by Hickmott in 1962 [49]. Hickmott studied five metal–oxide–metal sandwiches such as Al–SiO_2–Au, Al–Al_2O_3–Au, Al–Ta_2O_3–Au, Al–ZrO_2–Au and Al–TiO_2–Au under direct-current-voltage, in which Al_2O_3 was investigated extensively. The switching times of these oxides are below 1 μs, indicating the excellent performance of oxide in resistive switching. Up to now, numerous kinds of SMO have been investigated. For example, the highest ON/OFF ratio ($>10^9$) was realized in the research of TaO_x and GeO_x devices [50, 51]. HfO_x and TaO_x based devices both reached nanosecond level in program speed [52–54]. ReRAMs based on other metal oxides such as CuO_x [55–57], CoO_x [58, 59], NiO_x [60], etc, have also been studied systematically. The underlying mechanism of resistive switching behavior of metal oxides is mainly put down to the formation and rupture of conductive filaments (CFs) or the changes of valence states caused by the migration of oxygen vacancy [61, 62]. For SMO-based ReRAM, owing to the production and the migration of oxygen vacancies inside metal oxide film, conductive filaments can evolve and resistive switching or memory function can be realized without active metal electrode and metal conductive filaments. Commonly speaking, 'electroforming' process is required when the first resistive switching happens in SMO-based ReRAM. In this process, some oxygen atoms inside oxides will lose electrons and oxygen vacancies will be produced under high applied voltage accompanied with generating O_2. And then the redistribution of these oxygen vacancies causes the changes of resistance states of the metal oxide film under different applied voltages, resulting in the memory function of ReRAM.

In conclusion, we have listed some major applications of SMO and discussed them in limited space, but we don't enumerate all the applications as they are extremely wide. In this book, more detailed discussion in SMOs and their application in different fields will be presented.

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Chapter 10

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Chapter 11

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