Strain Engineering in Thin Films

A thesis submitted in partial fulfilment of the requirement
For the award of degree in
Master of Science
in
Physics

Submitted by
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CERTIFICATE

This is to certify that the report entitled **"Strain Engineering in Thin Films"**, submitted by Jyoti Mahajan, Roll No. 301904005, in partial fulfilment of requirements for the award of degree M.Sc. in Physics from School of Physics and Materials Science, Thapar Institute of Engineering and Technology, Patiala is a record of candidate own work carried out by her under my supervision and guidance. The work reported here has not been submitted, either in part or in full, for the award of any other degree in other institute or university.

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DECLARATION

I hereby declare that the thesis report entitled **"Strain Engineering in Thin Films"** submitted by me in partial fulfilment of the requirements for the award of degree of Master of Science in Physics submitted in the School of Physics and Material Science at Thapar Institute of Engineering and Technology, is an authentic record of my work carried out under the supervision of **Dr. Bhaskar Chandra Mohanty** and refers other researcher`s work which is duly listed in the reference section. I further declare that work embodied in this report has not been and will not be submitted, either in part or in full, in any other institute or university for award of master and science or any another other degree.

Jyoti

Jyoti Mahajan

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ABSTRACT

Strain engineering for improving properties of thin films, and hence performance of thin film based devices has drawn significant research interest. It has found technological relevance, especially in flexible devices that have broad coverage for future applications. The flexible devices need to survive mechanical distortion such as bending, twisting and stretching without losing its performance. Transparent devices must retain their transparency despite these operations. Strain engineering has been shown to improve fracture behaviour and tailor optical band gap of the inorganic semiconductor thin films. A brief literature study is presented in order to understand the strain and its effects on thin films.

CONTENTS

CHAPTER 1
INTRODUCTION

1.1 Thin films

Conventionally, thin film corresponds to a layer of material having thickness in the range of a few nanometres to hundreds of micrometres [1]. In such a form of material, the number of atoms on the surface compared to those in the bulk is very high. Owing to this very high ratio of surface to volume atoms, thin films exhibit properties very different from the bulk counterpart [2]. In addition to the exotic properties of the thin films, the growing trend of miniaturization of devices that requires minimum use of materials has led to extensive research and development in thin film technology. In fact, over the years, thin films have found increasing use in a variety of applications like microelectronics, flat-panel displays, energy generation and storage, computer chips, solid-state lighting, hard and protective coatings, micro-electrochemical system, automobiles, sensors etc. [3-5].

The deposition of thin films is a self-organising development that depends on the arrangement of discrete atoms/molecules on a substrate. This arrangement of depositing species, as expected, determines the eventual quality and properties of the films. There are varieties of deposition techniques, each having its own advantages over the others [6]. The choice of a deposition technique for a specific thin film mainly lies on how easily good quality films with desired properties can be routinely prepared at low cost. Nevertheless, in each deposition process there lie a number of process parameters such as growth rate, temperature, etc. that strongly influence the properties of the films. In the hindsight, it provides an opportunity to manipulate the properties of the films just by altering certain process parameters. The fabrication of films with easy-tunability of properties has become a key technological issue.

As far as manipulation of properties of the films is concerned, strain in the films plays a critical role [7]. In fact, strain has been shown to alter mechanical and opto-electronic properties. In the recent years, there have been efforts to identify process parameters that affect the strain in the films, which in turn helps in manipulating properties of the films. The intentional manipulation of strain or the so called ‘strain engineering’ as a tool to tailor properties of films has attracted significant research interest in recent years [8-9]. Especially, in flexible optoelectronic devices, strain engineering has been found very useful to change band gap of the films. On the other hand, strain engineering in thin films grown on flexible substrates is easily achieved because of tolerance of flexible substrates to mechanical distortion that yields strain in the films. This report deals with the strain engineering in the thin films and highlights some of the studies dealing with it.

1.2 Strain in thin films

Stress and its effects are traditionally considered to arise from external applied forces. However, in thin films there is some stress present even when apparently any external force is absent. This is known as residual stress in thin films. It affects various phenomena such as formation of film surface, generation and growth of crystalline defects, adhesion of film to the substrate, etc. On the other hand, during growth of thin films, stress usually arises from the thermal mismatch difference between substrate and films, film growth parameters, film thickness, post-deposition heat treatment, inclusion of impurities such as doping [10], and due to the type of substrate, [11] which helps in knowing the orientation and quality of films [12-17]. Nevertheless, substrate distortion and deformation also appear from stresses in the overlying thin films [18].

When a stress is applied to a crystalline solid, it can strongly perturb its electronic structure by changing the crystal symmetry or inducing unexpected defect which can modify the material intrinsic properties. Since, there is a correlation of the existence of strain with mechanical parameters such as fracture energy, hardness, critical strain, modulus and film strength, mechanical distortion of substrate can be a potential cause of strain in thin films. Although the existence of residual stresses is usually unacceptable, there are certain cases when it becomes advantageous. The presence of strain in films will open new application fields due to the modified properties. Conversely, flexible devices are evaluated for their reliability based on their tolerance to mechanical deformations such as twisting, bending, folding and stretching [19-21].

Figure1.1 shows deformation of the films relative to the substrate because of misfit epitaxial growth and surface tension. The film having internal tensile stress will tend to bend the substrate concavely upward and on the other hand internal compressive stress in the thin film will bend the substrate convex outwards. This helps in determining the mechanism that causes films to shrink to stretch relative to the substrate [18].

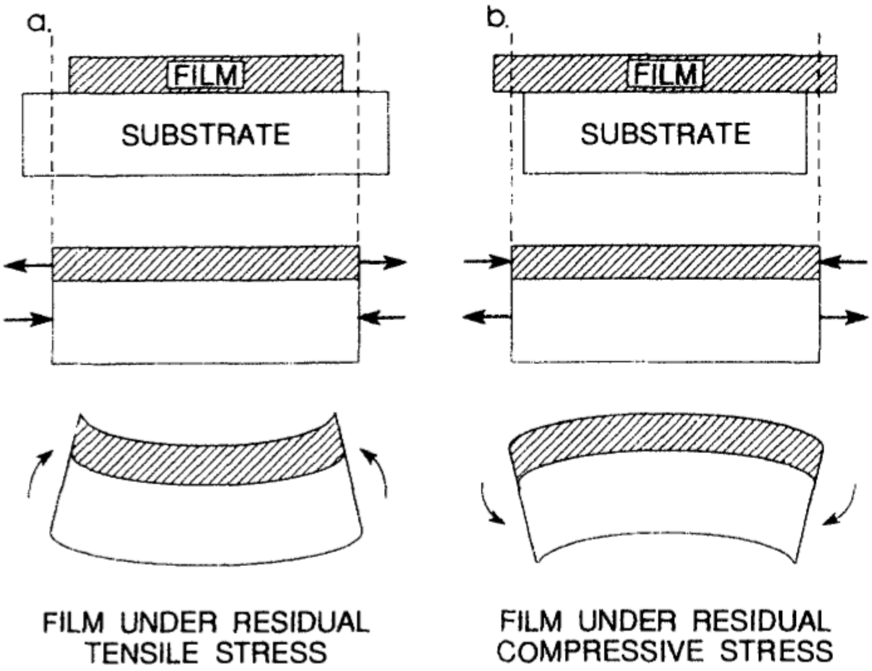

Fig1.1: Arrangement of events leading to (a) residual tensile stress in films and (b) compressive stress in films. [18]

1.3 Strain engineering in thin films

Strain engineering is regarded as an effective method to tailor optical, magnetic, electric, and mechanical properties of inorganic thin films [22-24].By inducing deformations, changes in electronic structure, extension or shrinkage of bond lengths[25-26], leading to the change in band structure of semiconducting films can be expected [25-30]. Because of the stress induced unintentionally in rigid thin films, there is a change in optical band gap via lattice and thermal substitution [31,32], thickness of the film, etc. [29-33]. On the other hand, the presence of impurities and defects causes stress inevitably in the thin films, which in turn changes its optical and electronic properties. Hence, by introducing the impurities, optical band gap can be modulated; therefore this ‘stress engineering’ is used in designing the electronic devices.

1.3.1 Flexible substrates for strain engineering

Strain engineering in thin films using flexible substrates have drawn significant interest in the last decade. Since earlier times, fabrication of electronic devices on flexible substrates has been in great demand in various fields of applications like sensors, robotics, displays etc.

Thus, these materials have crucial role in the advancement of devices. With the use of flexible material, there is decrease in the weight of flat panel displays. It provides ability to bend, conform and roll a display into different shapes. Among the typical flexible substrates namely ultrathin, metal foil and plastic substrates, the plastic substrates are light weight, have greater flexibility, and are less expensive. This makes plastic foils a great choice for the use as substrates in devices. In thin film technology, polyethylene terephthalate (PET), polyethersulfone (PES), polyimide (PI), polyethylene naphtha late(PEN) are prominently used due to their good thermal stability, dielectric and mechanical properties.

1.3.2 Substrates for use in flexible electronic devices

Selection of substrate is also very important parameter for the film growth. Matching in crystal growth and lattice parameter will strongly affect the growth behavior. The lattice mismatch between substrate and film induces strain of various kinds with varying degrees. Owing to their properties, PET and PES have been considered to be promising substrates for flexible optoelectronic devices. The typical properties of PET and PES are listed in Table 1.1.

Table 1.1: Properties of PET and PES substrates

Properties	**PET**	**PES**
Density (x10^3 kg/m^3)	1.375	1.37 – 1.46
Mechanical properties		
Tensile Strength (MPa)	55-75	67.6 – 89.6
Yield strength (MPa)	60-80	84.1 - 89.6
Elongation (%)	50-100	60- 80
Young modulus (E) (GPa)	2.8 - 3.1	2.41 - 2.83
Thermal properties		
Melting Point (°C)	260	150-180
Transition temperature (°C)	67–81	~225
Thermal Expansion (10^{-6}/K)	20-80	275.15-350

CHAPTER 2

LITERATURE SURVEY

Deposition of thin films on plastic substrates has been of growing interest because of its many applications in flexible optoelectronics. Although there have been many physical and chemical routes to prepare films on the plastic substrates, many of the functional layers are brittle in nature. Therefore, they can withstand limited stress arising due to mechanical distortion such as bending, stretching or twisting. This chapter highlights some of the studies that deal with improving mechanical strength of the films and strain engineering affecting various properties of the films.

2.1 Effect of strain engineering on fracture behaviour

Choi et al. (2015) have demonstrated the beneficial impact of strain engineering on fracture behaviour of Al doped zinc oxide (ZnO: Al) thin film deposited by RF magnetron sputtering [34]. The summary of the work is presented in Fig. 2.1.They have carried out strain engineering by depositing the films on a pre-bend PES substrates. The pre-bending of the substrate induced tensile strain. After deposition of the films, the substrates were released from the bending position and were made flat. They observed the 70% advancement in the fracture behaviour of Al doped zinc oxide (ZnO: Al) thin film by this method. It was observed that pre-bending of radius of curvatures equals to 14.3, 10.1and 8.2 mm created different amounts of tensile strains (0.70, 0.99 and 1.21%, respectively).On releasing it from the bent position to normal position residual compressive stress in the deposited film. The authors investigated the impact of this pre-bending strain in the fracture behaviour of the films. During a bending test, initiation of cracks perpendicular to the bending direction, saturated crack density and crack spacing were studied. It was found that cracks appeared at a lower bending strain for the films deposited on flat surfaces. However, films deposited on pre-bend substrates could withstand a larger bending strain [34].Hence; it depicts the positive influence of the residual compressive stress.

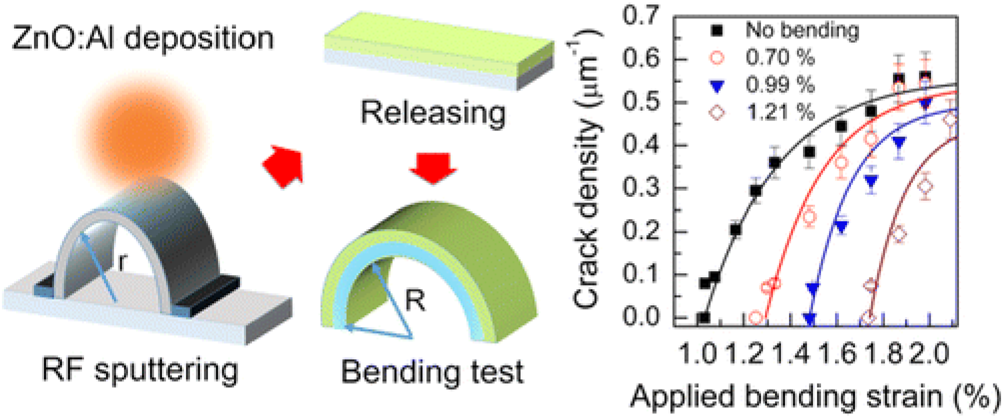

Fig. 2.1: The films were deposited on a pre-bend substrate. The substrate was released from its bending position after deposition of the film. The pre-bending radius allowed to incorporate specific amount of strain in the films. The films were subjected to a bending test and the results are shown in the third panel [34]

In a similar study Kim et al (2019) explored effects of strain engineering in PbS thin films grown by chemical bath deposition technique [13].With additional compressive stress, the films exhibited~ 18% improvements in the crack initiating strain and ~121% improvement in the fracture energy. Inducing additional stress in the thin films was achieved by depositing films on either convex or concave surface of a pre-bent PET substrate. The deposited film on a convex surface possessed compressive stress while on a concave surface it exhibited tensile stress when the substrate was released from bending mode to flat position [35-37]. Figure2.1 represents the evolution of crack density with applied bending strain for thin film deposited at different pre-bending strain. Bending curvature gradually increases from 0 to 1.3%, with increase in the crack density. On the other hand, for -0.88% compressive stress, it exhibited highest critical strain (0.77%), thus suggesting improved tolerance of the films to mechanical distortion during the bending test [38].

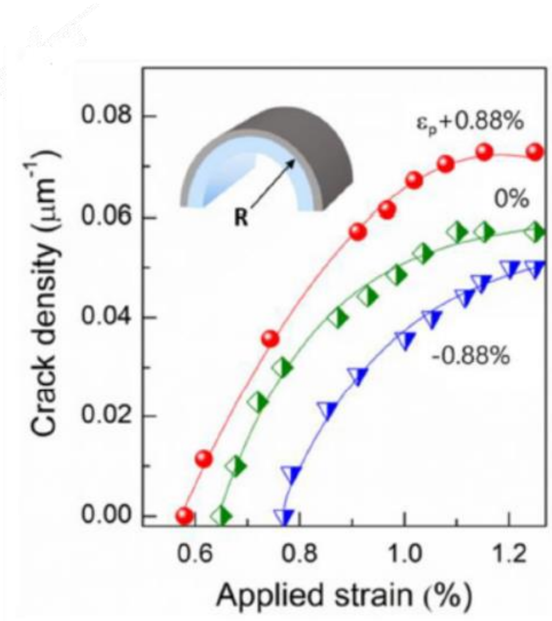

Fig. 2.2: Representation of crack density vs. applied bending strain with in-situ pre-bending strains of 0.88%, 0%, and 0.88% [37].

Cho et al (2015) presented a new way in the improvement of compressive residual stress in a thin film by stretching a PES substrate. The schematic of deposition of the film is shown in Fig. 2.3.

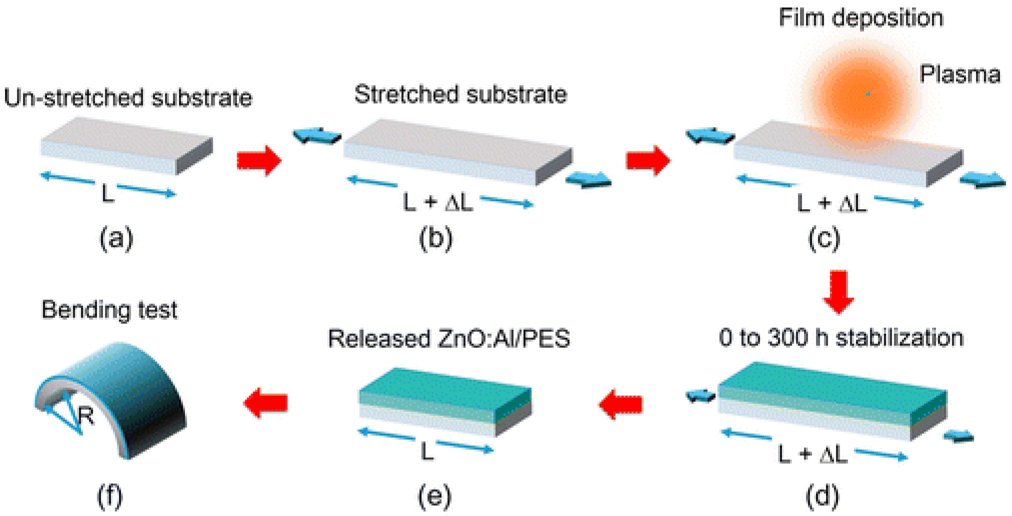

Fig. 2.3: Schematics of deposition process steps on a stretched polymer substrate [39]

In this ZnO thin film was deposited on a stretched PES substrate of 200 μm thicknesses. It was intentionally stretched with values ranging from 0.495 to 4.87% and stabilized for 300 hours. On releasing the film after the stabilization process, it is observed that with increase in the strain value from 0.49% to 4.87%, there is increase in the number of cracks while crack spacing starts decreasing [39]. As the stabilization process increases from 100 h, then there is an increase in the critical strain from 1.5% to 1.83% as compared to the reference sample. The results are summarized in Fig. 2.4. It implies that there will not be crack formation and hence, films can hold for greater bending strain.

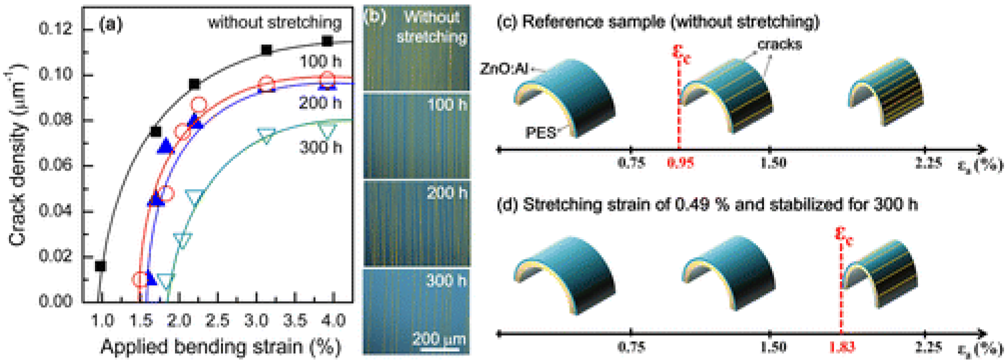

Fig. 2.4: Impact of strain engineering on fracture behaviour of the ZnO: Al thin films [45]

Moghal et al (2012) investigated mechanical properties of AlO_x thin films grown by different sputtering configuration (direct current (DC) magnetron sputtering, radio frequency (RF) magnetron sputtering and high-power impulse magnetron sputtering (HiPIMS)) on identical substrates [40]. The fragments tests revealed that by stretching the substrate under unaxial tension, the films cracked and the crack density increased with the increasing strain level. Among all of these, DC sputtering technique showed wide gap between the cracks at 4.08% critical strain, while RF and HiPIMS have 2.93%, 6.79% respectively[40]. The films grown by DC magnetron sputtering has the highest critical onset strain indicating greatest cohesive strength of oxide layer.

Zhang et al (2010) investigated the effect of residual stress in the SiO_x film of varying thicknesses on the strength, fracture energy and crack densities [41]. The amount of residual stress was evaluated by using a curvature method using multiple cracking analyses. Multiple film cracking (MFC) is performed on SiO_x ($x=1.7$) film of different thicknesses (43, 67, 90, 120, 320nm) deposited on PET substrate of thickness 12μm [41].With increase in thickness, the amount of residual stress was found to decrease. As the compressive residual stress in the film increased, there was increase in the fracture energy, and film strength and interfacial shear strength (IFSS) of the specimen.

2.2 Effect of strain engineering on optical bandgap

Lee et al (2018) proposed that on inducing intentional tensile or compressive stress in a flexible PbS thin film, the band edge positions and optical band gap can be modulated [42]. The film having thickness ~200nm were grown on a pre-bent PET substrate at 40°C in the chemical bath deposition method. After deposition of the films, the specimen was released back to the initial position (i.e., flat position). The specimen gave a concave or convex shape to the substrate at different curvatures [42]. The pre bending of the substrate with certain radii is intended to deliberately introduce a stress into the film by releasing the bending position to the flat one. The unstrained PbS film (i.e., the one deposited on the flat substrate) was found to have an optical band gap of 1.62ev [43]. On the other hand, pre bending during deposition of the film and subsequent release of the substrate significantly affected the band gap of the films. For instance, an increase in band gap to1.73eV for 0.80% pre-bending strain and a decrease to a value of 1.43eV for -0.88% pre-bending strain were observed. This, modulation of band edges will further help in potential development of flexible optoelectronics.

Choi et al. (2018) have studied the modulation in optical band gap of ZnO thin films by deliberate introduction of strain in the films grown on PET substrates [44]. The substrates were stretched and the film was deposited by RF magnetron sputtering. After deposition, the substrates were released that introduced strain the films. It was shown that the optical band gap can be modulated by about 150 meV depending upon the amount of strain introduced in the films. These results showed how the band gap of the films can be intuitively manipulated by strain engineering.

In another work (2019), the same group investigated how deposition of ZnS thin films on pre-stretched PET substrates can lead to introduction of crystal anisotropy and modulation of band gap [45]. They grew ZnS films by magnetron sputtering on stretched PET substrates. After the depositions, the substrates were held in that stretched position for about 6 hr and then were released. It was found that by stretching to different degrees, strains of varying amounts were introduced. This strain modulated the refractive index of the samples. Figure 2.5 shows the variation of refractive index for ZnS thin film at different strain values. There was a reduction in the optical band-gap ~110meV of a thin film and refractive index was decreased to 2.05 on applying intentionally compressive strain ~ 4.87% [45].

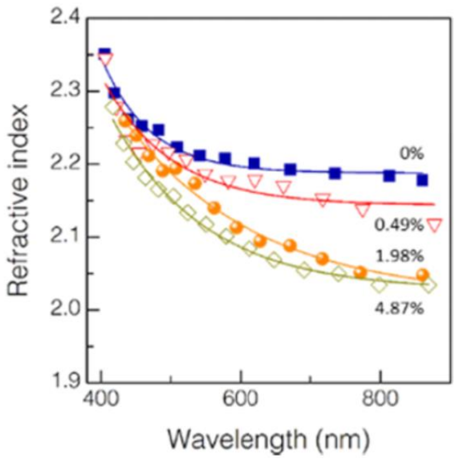

Fig 2.5: Variation in the refractive index of the film versus wavelength for highly strained samples [45].

Mohanty et al. (2009) have investigated the effect of stress arising due to change in thickness of ZnO:Al thin films and its effect on optical band gap. They have shown that there was an in-plane compressive stress due to bombardment the energetic depositing particles on growing surface of the film. As the thickness of the films increased, the stress in thin film became less compressive due to which the band gap changed.

Hsieh et al. (2012) determined the effect of strain on ZnO:Al and indium tin oxide (ITO) films [46]. The films were deposited on pre-strained flexible substrates of different thicknesses. It is seen that with decrease in the radius of curvature, the strain increased. For similar thickness of ZnO:Al and ITO films, rate of deposition for the ZnO:Al was lower. This gave better results of mechanical properties for pre-strain and hence, can tolerate more stress than ITO film[46].

The optical properties of the SnO_2 films were examined by Zhou et al. (2014)[47]. They prepared epitaxial SnO_2 thin films of different thicknesses (30 - 130 nm) on polished Al_2O_3substrates by RF sputtering. It was observed that with the increase in the thickness, the tensile strain induced by the mismatch between Al_2O_3 substrate and SnO_2 thin film was reduced [47]. The band gap of the films did not show a linear relationship with the film thickness. The band gap reduced as high as by 0.9 eV with decreasing film thickness.

Liu et al (2015) examined the shift of band gap in Ga – doped ZnO thin films. The films of varying thicknesses were deposited on glass substrates by magnetron sputtering [48]. It was observed that the band gap linearly depended on the in-plane stress and the electron concentration. For films in compressive stress, narrowing of band gap was observed while for tensile stress the band gap was widened [48].

CHAPTER 3

SUMMARY

It is summarized that strain in thin films plays a crucial role in mechanical and optical properties of the films. Thus, by artificially manipulating the strain in the film one can tailor the properties suitable for specific applications. Certain studies have revealed that by intuitive design of deposition configuration, for example deposition on a pre-stretched or pre-bent substrate, can help in tuning properties of the films. Considerable improvement in fracture tolerance in thin films have been obtained for the films which were subjected to strain engineering. This shows high technological implication of strain engineering, especially in the development flexible devices.

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