

Reviewing Optical Properties of Chalcogenide Glass Solution-Driven Thin Films

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ABSTRACT

Chalcogenide glasses (ChGs) have gained significant attention in recent years due to their unique optical properties and potential applications in various fields such as photonics, infrared sensors, and telecommunications. Among the different forms of ChGs, solution-driven thin films have emerged as a promising avenue for exploring their optical properties and enabling novel device designs. This research paper provides a comprehensive review of the optical properties of Chalcogenide glass solution-driven thin films, highlighting their synthesis methods, characterization techniques, and potential applications. The paper also discusses the key factors affecting the optical properties of these films and presents recent advancements and challenges in the field.

Keywords: Chalcogenide glasses, Photonics, Infrared Sensors, and Telecommunications

INTRODUCTION

Chalcogenide glasses (ChGs) are a class of amorphous materials composed of chalcogen elements (sulfur, selenium, tellurium) combined with elements such as arsenic, antimony, or germanium. ChGs exhibit unique optical properties, making them attractive for a wide range of applications in photonics, infrared optics, and telecommunications. The combination of their low phonon energy, high refractive index, and strong infrared transparency makes ChGs particularly useful for the development of devices operating in the mid-infrared (mid-IR) spectral range.

Importance of Thin Films in Optical Applications:

Thin films play a crucial role in various optical applications. They offer several advantages over bulk materials, including the ability to control film thickness and uniformity, compatibility with miniaturized device designs, and the possibility of tailoring optical properties through film composition and structure. Thin films also enable the integration of ChGs with other materials and structures, expanding their functionality and application potential. In optical devices such as waveguides, modulators, and sensors, thin films offer enhanced light-matter interactions and allow for the efficient manipulation and control of light.

Motivation for Studying Chalcogenide Glass Solution-Driven Thin Films:

The exploration of ChG solution-driven thin films has gained significant attention due to several compelling reasons. Firstly, solution-based techniques offer a cost-effective and scalable approach for the deposition of ChGs on various substrates, enabling large-area film production and compatibility with fabrication processes such as roll-to-roll manufacturing. Secondly, solution-driven thin films provide the ability to tune the composition and stoichiometry of ChGs, thereby tailoring their optical properties for specific applications. Additionally, solution processing techniques allow for the integration of ChGs with other functional materials, facilitating the development of hybrid and composite structures with enhanced optical performance. The study of ChG solution-driven thin films holds the promise of advancing the understanding of their optical properties, expanding their application potential, and driving the development of next-generation optical devices.

SYNTHESIS METHODS FOR CHALCOGENIDE GLASS SOLUTION-DRIVEN THIN FILMS:

Several synthesis methods have been developed for the deposition of ChG solution-driven thin films. These techniques offer versatility in terms of film thickness, composition control, and compatibility with different substrates. Some common methods include:

Spin coating: In spin coating, a liquid ChG precursor solution is dispensed onto a rotating substrate, causing the solution to spread and form a thin film due to centrifugal force. The film is subsequently dried and thermally treated to remove solvents and promote ChG formation.

Dip coating: Dip coating involves immersing a substrate into a ChG precursor solution and then withdrawing it at a controlled rate. The withdrawal speed determines the film thickness, and subsequent heat treatment converts the precursor film into a ChG film.

Sol-gel methods: Sol-gel techniques utilize hydrolysis and condensation reactions of metal alkoxides or metal salts to form a precursor sol, which is then deposited onto a substrate by spin coating, dip coating, or other techniques. Subsequent thermal treatment leads to the formation of ChG thin films.

Vapor deposition techniques: Vapor deposition methods, such as thermal evaporation or sputtering, involve the evaporation or deposition of ChG materials from a solid source onto a substrate. These techniques allow for precise control of film thickness and uniformity but require specialized equipment. The choice of synthesis method depends on factors such as desired film thickness, substrate compatibility, and film quality requirements. Each technique offers its advantages and limitations, and researchers continue to explore and optimize these methods to achieve high-quality ChG solution-driven thin films.

Molecular Precursor Strategies:

In addition to solution-based techniques, molecular precursor strategies have emerged as effective approaches for depositing ChG solution-driven thin films. These strategies involve the synthesis and utilization of precursor molecules that can undergo chemical reactions to form ChGs upon heat treatment. Molecular precursor strategies offer advantages such as precise control over film composition, improved film quality, and the ability to incorporate dopants or functional groups into the ChG matrix. Some commonly employed molecular precursor strategies include:

Metal-Organic Chemical Vapor Deposition (MOCVD): MOCVD involves the pyrolysis of metal-organic precursors in a gaseous phase to deposit ChG films onto heated substrates. Metal-organic precursors containing chalcogenide ligands undergo decomposition, releasing metal atoms and chalcogen species that react to form ChG films.

Chemical Vapor Deposition (CVD): CVD techniques utilize volatile chalcogenide precursors, typically in combination with metal precursors, which react at elevated temperatures to deposit ChG films. CVD offers precise control over film composition and stoichiometry by adjusting the precursor flow rates and reaction conditions.

Vapor Deposition Techniques (e.g., Thermal Evaporation, Sputtering):

Vapor deposition techniques, including thermal evaporation and sputtering, have been widely employed for the fabrication of ChG thin films. These techniques involve the deposition of ChG materials in a vapor phase onto a substrate, forming thin films through condensation or chemical reactions. Key vapor deposition techniques include:

- In thermal evaporation, a ChG material is heated to its vaporization temperature in a vacuum chamber. The evaporated ChG material condenses onto the substrate, forming a thin film. The film thickness can be controlled by adjusting the deposition time or the evaporation rate.
- Sputtering is a physical vapor deposition technique that involves bombarding a ChG target material with energetic ions in a low-pressure gas environment. The collision of ions with the target ejects ChG atoms, which then deposit onto the substrate, forming a thin film. Sputtering allows for precise control over film thickness and composition and is compatible with various substrate materials.

Both thermal evaporation and sputtering techniques offer advantages such as high film purity, good control over film thickness, and compatibility with large-scale manufacturing processes. However, they require specialized equipment and high vacuum conditions for deposition. The choice of molecular precursor strategies or vapor deposition techniques depends on factors such as film quality requirements, desired film composition, and the available equipment. Researchers

continue to explore and optimize these methods to achieve ChG solution-driven thin films with improved optical properties and performance.

THE CHEMISTRY OF CHALCOGENIDES IN SOLUTION

Mechanisms of Dissolution

There are many important implications that may be drawn from an understanding of the mechanism of chalcogenide dissolution. To begin, understanding the solution and the films produced through that solution requires a thorough understanding of how glass dissolves. Second, gaining this kind of insight into the basic sciences helps shed light on how to enhance current manufacturing procedures and add new materials to their repertoire. As such, this section details the chemistry behind arsenic sulfide's dissolution in various solvents, and compares it to that of other chalcogenides. Arsenic sulphide solubility in n-propylamine and n-butylamine was studied by Chern et al. The researchers assumed that the amorphous arsenic sulphide used as a starting material would have a layered structure and postulated that the solvent would break the material down into little flat clusters, first at defect sites between the layers. This takes place when an alkyl amine group from the solvent replaces a sulphide atom in a compound. The ensuing arsenic alkyl ammonium group loses a hydrogen atom to produce an RNH³⁺ group, which subsequently forms a covalent link with the sulfur's lone negative bond. Due to the extremely electronegative nature of nitrogen, the arsenic atom in the alkyl ammonium group now has a lower electron density than it did before the substitution. As a result, a molecule containing alkyl amino arsenic and three hydrogen ended sulphide groups can form after a second and third nucleophilic assault on the arsenic. The chemical balance between the hydrogen sulphide groups and the alkyl ammonium sulphide salt is maintained by the presence of additional solvent molecules. When arsenic sulphide layers are shattered, an insoluble alkyl amino arsenic complex precipitates together with amorphous, arsenic-deficient arsenic sulphide pieces terminated by excess sulphide dangling bonds and charge compensating alkyl ammonium ions (Fig. 1). The size (and form) of the arsenic sulphide fragments (on the order of 2-10 nm) influences the magnitude of the sulphur excess. Indeed, Kohoutek et al. showed that the concentration of arsenic sulphide in n-butylamine correlates with the cluster size of the dissolved arsenic sulphide. The arsenic sulphide fragments are protected by a polar alkyl ammonium sulphide shell, making them more soluble in the solvent of choice and a wide variety of other polar solvents. Since the precipitating alkyl amino arsenic molecule is likely a mixture of diverse reaction products, including oxide-species from a competing nucleophilic attack by trace amounts of water in the solvent, its specific composition or even the chemical formula could not be identified. Arsenic sulphide dissolution products in n-propylamine, however, would be as follows from a thermodynamic (ideal, in chemical equilibrium) standpoint:

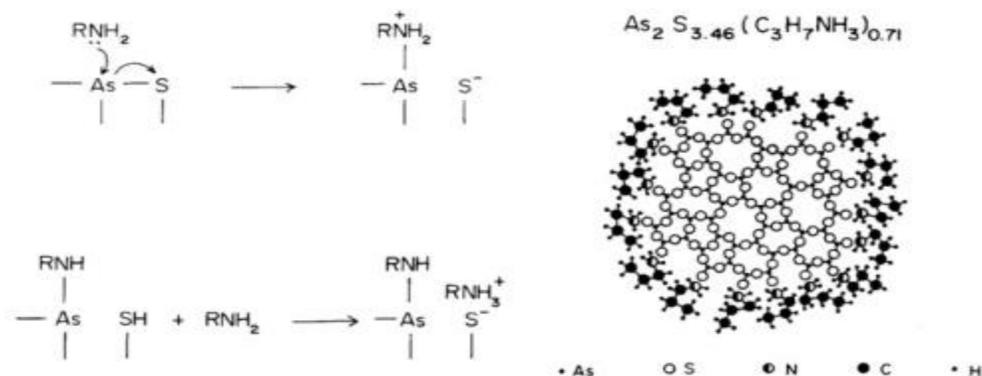
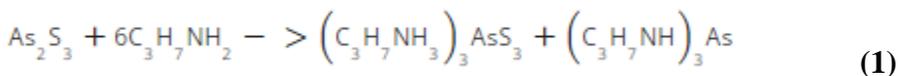


Fig.1 : Chemical equations and cluster image. (Left) Electrophilic substitution reaction proposed for the arsenic sulfide dissolution process. (Right) Proposed schematic structure of As₂S₃ + n-propylamine. Reprinted with permission from J. Appl. Phys. 54, 2701(1983).



For a low-temperature route to optical chalcogenide glass films, Guiton and Pantano explored the dissolution of arsenic sulphide in ethylenediamine (EDA) in the late 1980s. Polymer-like chains of As₄S₄ rings interlinked by bridging sulphur atoms, each ring chelated by two solvent molecules, were proposed as an alternative to the generation of alkyl ammonium salts or hydrogen sulphide in this system. The chelating properties of a diamine solvent are what make this alternative method viable. Similar to the sol-gel process used to create silicate gels (Fig. 2), the material condenses as the solution evaporates. According to Guiton et al., the absence of alkyl ammonium species in their water-free solutions was due to the fact that they lacked a proton source (other than the necessary amino group) for their synthesis. Note, however, that Chern et al. found alkyl ammonium salts, despite the fact that their proposed dissolution mechanism does not need the presence of water molecules.

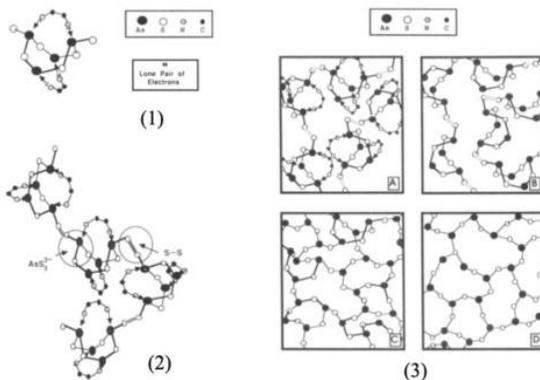


Fig. 2: Proposed As₂S₃/EDA solution species and gel-to-glass transition. (1) Lewis acid-base chelation model (2) Branched chains (3)(A) solution species; (B) amine loss; (C) elimination of As-S-S-As; (D) amorphous network. Reprinted with permission from Chem. Mater. 1(5), 558-563(1989).

CHARACTERIZATION TECHNIQUES FOR OPTICAL PROPERTIES

Characterization Techniques for Optical Properties:

To study the optical properties of ChG solution-driven thin films, various characterization techniques are employed. These techniques provide valuable insights into the film's transparency, reflectivity, absorption, photoluminescence, crystallinity, surface morphology, and structural properties. Some commonly used characterization techniques for optical properties of ChG solution-driven thin films include:

Optical Spectroscopy (Transmission, Reflection, Absorption):

- Measures the amount of light transmitted through the film as a function of wavelength. It provides information about the film's transparency, refractive index, and absorption properties.
- Measures the intensity and wavelength dependence of light reflected from the film surface. It helps determine the film's reflectivity and optical constants.
- Measures the amount of light absorbed by the film as a function of wavelength. It provides insights into the film's bandgap, absorption edge, and absorption coefficients.

Ellipsometry and Spectroscopic Ellipsometry:

Ellipsometry measures changes in the polarization state of light reflected or transmitted through the film. By analyzing the ellipsometric parameters, such as the phase shift and amplitude ratio, the film's thickness, refractive index, and optical properties can be determined.

Spectroscopic Ellipsometry extends ellipsometry to a range of wavelengths, enabling the characterization of thin film optical properties over a broad spectral range.

Photoluminescence Spectroscopy

Photoluminescence spectroscopy measures the emission of light from the film upon excitation with photons. It provides insights into the film's electronic structure, defect states, and luminescent properties. Photoluminescence spectroscopy can reveal information about the presence of impurities, dopants, or luminescent centers within the film.

Raman Spectroscopy:

Raman spectroscopy probes the molecular vibrations and lattice vibrations within the film by measuring the inelastic scattering of light. It provides information about the film's chemical bonding, structural properties, and crystallinity. Raman spectroscopy can identify the presence of different ChG phases and detect structural changes induced by processing or doping.

X-ray Diffraction (XRD):

XRD measures the scattering of X-rays by the film's crystal lattice, providing information about its crystalline structure, crystallographic orientation, and phase composition. XRD can determine the presence of crystalline phases in ChG films and quantify their crystallinity and grain size.

Scanning Electron Microscopy (SEM) and Atomic Force Microscopy (AFM):

SEM and AFM techniques provide high-resolution imaging of the film's surface morphology, topography, and roughness. SEM offers three-dimensional information, while AFM allows for nanoscale surface characterization. These techniques are crucial for assessing film quality, uniformity, and the presence of defects or surface features that can impact the film's optical properties.

These characterization techniques provide complementary information about the optical properties, crystallinity, morphology, and structural properties of ChG solution-driven thin films. By utilizing these techniques, researchers can gain a comprehensive understanding of the film's behavior and optimize its optical performance for specific applications.

FACTORS AFFECTING THE OPTICAL PROPERTIES OF CHALCOGENIDE GLASS THIN FILMS

Composition and Stoichiometry:

The composition and stoichiometry of ChG thin films significantly influence their optical properties. Varying the chalcogenide element (e.g., sulfur, selenium, tellurium) and the other constituent elements (e.g., arsenic, antimony, germanium) alters the bandgap, refractive index, and optical transparency of the films. Modulating the composition allows tailoring the optical properties for specific applications, such as infrared sensing or integrated photonics.

Film Thickness and Morphology:

The thickness and morphology of ChG thin films affect their optical behavior. Film thickness influences the interference effects, light absorption, and transmission properties. Thin films with precise control over thickness are crucial for achieving desired optical performance, such as antireflection coatings or waveguides. Morphology, including grain size, surface roughness, and uniformity, can impact light scattering, transmission, and surface plasmon interactions, thus affecting the overall optical properties.

Annealing and Post-Processing Effects:

Annealing and post-processing steps after film deposition can significantly alter the optical properties of ChG thin films. Annealing treatments enable structural relaxation, recrystallization, and removal of defects, leading to changes in refractive index, crystallinity, and absorption characteristics. Controlled annealing conditions can enhance film quality and improve optical performance. Additional post-processing techniques like thermal treatment, ion implantation, or surface functionalization can also modify the film's optical properties and introduce tailored functionalities.

Substrate Selection and its Influence on Film Properties:

The choice of substrate material and its interaction with the ChG thin film can influence optical properties. Substrates with different refractive indices can modify film thickness, transmission, and reflectivity due to optical interference effects. The lattice mismatch and thermal expansion coefficient mismatch between the film and substrate can affect film quality, strain, and crystal structure. Proper substrate selection and optimization are crucial to achieve high-quality ChG thin films with desired optical properties.

OPTICAL PROPERTIES AND APPLICATIONS

Chalcogenide glass (ChG) thin films exhibit excellent optical transparency in specific wavelength ranges, making them suitable for various applications. The transparency of ChG films refers to their ability to transmit light without significant absorption or scattering. By carefully selecting the composition, ChG thin films can achieve transparency windows in the visible, near-infrared (NIR), and mid-infrared (MIR) spectral regions.

The optical transparency of ChG thin films enables their use in optical coatings, anti-reflection coatings, and windows for optical devices. These films can efficiently transmit light within specific wavelength ranges, allowing for the manipulation and control of light in photonic systems.

The refractive index of ChG thin films determines the speed at which light propagates through the material. ChG films typically exhibit higher refractive indices compared to traditional oxide-based materials. The refractive index can be tailored by adjusting the composition and stoichiometry of the ChG film.

The dispersion of refractive index refers to the dependence of refractive index on the wavelength of light. ChG thin films can exhibit significant dispersion, enabling the design of optical components with wavelength-dependent behavior, such as dispersion compensation elements and chromatic dispersion-based devices.

ChG thin films possess remarkable nonlinear optical properties, making them attractive for nonlinear optics and photonics applications. Nonlinear optical effects, such as second-harmonic generation, third-harmonic generation, and four-wave mixing, can be observed in ChG thin films.

These nonlinear optical properties arise from the unique electronic structure and strong intrinsic nonlinearities of ChG materials. Harnessing these properties allows for the development of nonlinear optical devices, including frequency converters, optical parametric oscillators, and all-optical switches.

ChG thin films find extensive applications in photonic devices and integrated photonics due to their unique optical properties. Some key photonic applications include:

- ChG thin films can be patterned into waveguides, which are structures that guide and confine light within a specific region. ChG waveguides exhibit low optical losses and can be integrated with other photonic components, enabling the development of compact and efficient optical circuits.
- ChG thin films are used in the fabrication of optical sensors for various applications, including chemical sensing, biosensing, and environmental monitoring. The high refractive index and sensitivity to changes in the surrounding medium make ChG films suitable for detecting minute changes in refractive index or absorption.
- ChG thin films can be utilized as active components in optical modulators, which control the intensity or phase of light. Their nonlinear optical properties enable efficient modulation of light signals, leading to applications in optical communications, data processing, and sensing systems.

ChG thin films excel in the infrared (IR) spectral region, making them valuable for a range of IR applications. Some notable IR applications include:

- ChG thin films are employed in thermal imaging systems, where they detect and convert IR radiation into visible images. The high IR transparency of ChG films in the MIR range allows for the detection of thermal signatures and the visualization of temperature variations.
- ChG thin films serve as optical elements in IR spectroscopy techniques. They can function as windows, lenses, or waveplates to transmit, focus, or modify IR radiation for spectroscopic analysis of materials in the MIR range. ChG films enable precise and accurate characterization of chemical composition and molecular vibrations.

These applications highlight the versatility of ChG thin films in photonics, nonlinear optics, and IR technologies. Their unique optical properties and tunability make them promising materials for a wide

RECENT ADVANCEMENTS AND CHALLENGES

Novel Fabrication Techniques and Strategies:

Researchers have been exploring various novel fabrication techniques and strategies to overcome challenges and enhance the optical properties of Chalcogenide glass (ChG) solution-driven thin films. Some of these techniques include:

Sol-gel Method: The sol-gel process involves the synthesis of ChG thin films from a precursor solution, followed by gelation and drying to form a solid film. This technique offers advantages such as low-temperature processing, good film uniformity, and the ability to incorporate dopants or nanoparticles into the film matrix.

Chemical Bath Deposition (CBD): CBD involves the deposition of ChG thin films from a solution containing chalcogenide precursors and a suitable reducing agent. It offers simplicity, cost-effectiveness, and the ability to deposit films on a variety of substrates.

Pulsed Laser Deposition (PLD): PLD is a physical vapor deposition technique that involves ablating a target material using a high-energy laser to deposit thin films onto a substrate. PLD enables precise control over film composition, stoichiometry, and thickness, allowing the fabrication of high-quality ChG films.

Enhanced Optical Properties through Alloying and Doping:

Alloying and doping are effective strategies to enhance the optical properties of ChG thin films. By incorporating different elements into the ChG matrix, researchers can modify the bandgap, refractive index, and optical transparency of the films. For example, alloying ChG materials with elements such as Ge or Ga can widen the transparency window and improve film quality.

Doping ChG films with rare earth elements, transition metals, or other dopants can introduce desired optical properties, such as enhanced photoluminescence or nonlinear optical behavior. Doping can also facilitate the tuning of refractive index and dispersion properties, enabling the design of advanced photonic devices.

Integration with Other Materials and Structures:

To expand the functionalities and applications of ChG thin films, integration with other materials and structures is essential. ChG films can be integrated with different substrate materials, such as silicon, polymers, or glass, to combine their optical properties with the advantages of the specific substrate. Integration with other materials, such as metals or dielectric layers, can enable the development of hybrid structures with enhanced optical properties, such as plasmonic effects, surface-enhanced Raman scattering (SERS), or improved light-matter interactions.

Challenges in Achieving Uniformity and Reproducibility:

Obtaining uniform and reproducible ChG thin films remains a challenge in their fabrication. Factors such as precursor stability, film nucleation and growth kinetics, and drying conditions can impact film uniformity and reproducibility. Controlling these factors and optimizing the fabrication processes are crucial to achieving consistent film quality and optical properties.

Stability and Reliability Considerations:

Stability and reliability are important considerations for ChG thin films, especially in practical applications. ChG materials are susceptible to environmental factors, such as moisture and temperature, which can degrade film quality and optical properties over time. Developing protective coatings or encapsulation strategies can enhance the stability and reliability of ChG thin films, making them suitable for long-term use in various applications.

In conclusion, novel fabrication techniques, alloying, and doping strategies have been explored to enhance the optical properties of ChG solution-driven thin films. Integration with other materials and structures enables the development of advanced photonic devices. Challenges such as achieving uniformity and reproducibility, as well as stability and reliability considerations,

need to be addressed to ensure the practical implementation of ChG thin films in optical applications.

CONCLUSION

Summary of the key findings and Insights

- The composition and stoichiometry of ChG thin films significantly influence their optical properties. Varying the chalcogenide element and other constituent elements allows tailoring the bandgap, refractive index, and optical transparency of the films for specific applications.
- Film thickness and morphology play a crucial role in the optical behavior of ChG thin films. Precise control over film thickness is essential for achieving desired optical performance, while morphology affects light scattering and transmission properties.
- Annealing treatments and post-processing techniques can modify the optical properties of ChG thin films. Annealing enables structural relaxation and defect reduction, leading to changes in refractive index and absorption characteristics.
- The choice of substrate material influences the optical properties of ChG thin films. Different refractive indices and lattice mismatches between the film and substrate can modify film thickness, transmission, and reflectivity.
- Alloying and doping ChG thin films offer opportunities to enhance their optical properties. Incorporating different elements into the ChG matrix or introducing dopants can widen the transparency window, improve photoluminescence, and enable nonlinear optical behavior.

Potential Future Directions for Research and Applications:

Based on the findings and insights presented in this research paper, several potential future directions for research and applications in the field of ChG solution-driven thin films can be identified:

- Further exploration of novel fabrication techniques, such as sol-gel methods or chemical bath deposition, can improve film uniformity and reproducibility, enabling large-scale production of high-quality ChG thin films.
- Investigating the integration of ChG thin films with other materials, such as metals or dielectric layers, can enable the development of hybrid structures with enhanced optical properties and functionalities. Exploring the integration of ChG films with emerging nanomaterials or 2D materials could also lead to new opportunities.
- Continued research on alloying, doping, and compositional engineering of ChG thin films can further enhance their optical properties. Exploring new combinations of elements, dopants, and stoichiometries can expand the range of optical functionalities and enable novel applications.
- Further development of ChG thin film-based photonic devices, such as waveguides, sensors, modulators, and photonic integrated circuits, can lead to advancements in telecommunications, sensing technologies, and data processing. Exploring ChG thin films for infrared applications, including thermal imaging and spectroscopy, can also yield significant benefits.
- Investigating stability and reliability aspects of ChG thin films, such as environmental durability and long-term performance, is crucial for practical applications. Developing protective coatings or encapsulation strategies can enhance the stability and reliability of ChG thin films.

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