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Luminescent Properties of Sulfide Glass-Ceramics: Mechanisms, Doping Regulation and Application Progress

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Abstract
Sulfide glass-ceramics have emerged as a research hotspot in high-efficiency luminescent materials due to the low phonon energy advantage of sulfide crystalline phases. They integrate the engineering merits of glass, such as easy forming and processability, with the excellent luminescent properties of crystals. Therefore, the systematic study of their related properties holds important scientific and application value for the development of optoelectronic devices. Based on a biphasic structure composed of glass and crystalline phases, the luminescent properties of these materials are jointly affected by crystallite size, glass-crystallite interface, crystalline phase purity and doping site distribution. They can be classified by the composition of crystalline phases, type of luminescent centers and excitation mode, with emission intensity, quantum yield and other parameters serving as the core evaluation indices for their luminescent performance. Their luminescence is divided into intrinsic and doped luminescence: the former is dominated by exciton and defect luminescence of CdS crystallites, which is regulated by the quantum confinement effect and defect types; the latter centers on rare earth, transition metal ion doping and quantum dot/rare earth composite doping, with site regulation, energy transfer, concentration quenching and crystalline phase structure regulation as the key optimization mechanisms that require the synergistic matching of multiple parameters. At present, such materials have exhibited application potential in the fields of near-infrared solid-state lighting, optical information storage, temperature sensing, white light illumination and so on, showing the developmental characteristics of being device-oriented, scenario-specific and capable of dynamic modulation. Meanwhile, the research still faces several challenges, including insufficient chemical stability of CdS-based materials, easy concentration quenching caused by high-concentration rare earth doping, environmental issues of PbS-based systems, and unclear interfacial energy transfer mechanism. Future research needs to focus on these key problems to promote the evolution of sulfide glass-ceramics from material exploration to high-performance device application.

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1. Introduction

Luminescent materials are being increasingly applied in fields such as information display, solid-state lighting, optical communication and optical storage, which imposes higher requirements on the comprehensive performance of matrix materials. Glass-ceramics integrate the structural units of both glass and crystals, and their research value lies in the achievable synergistic optimization of structure-performance through controlled crystallization. Zi *et al*. proposed that the combination of a transparent

glass matrix and functional nanocrystalline phases constitutes an effective approach to developing integrated material systems with luminescent and photoresponsive properties [1]. This finding indicates that the continuous network provided by the glass phase not only facilitates large-size forming, uniform component dispersion and subsequent processing, but also enables the regulation of the type, size and distribution of crystalline phases via heat treatment, thus endowing the materials with designable luminescent behaviors. For glass-ceramics designed for luminescence, this biphasic composite characteristic forms an important foundation that distinguishes them from single-phase glass or crystals.

Among various glass-ceramic systems, the migration of luminescent centers to low phonon energy environments is recognized as a crucial direction to improve radiative efficiency. Liang *et al*. found that the upconversion luminescence intensity and lifetime of rare earth ions are significantly enhanced after the precipitation of low phonon energy crystalline phases [2]. This demonstrates that crystalline phases can provide a more favorable local coordination environment for luminescent ions than glass networks, and reduce the non-radiative loss caused by multiphonon relaxation. Sulfide glass-ceramics have attracted extensive attention due to the low phonon energy of their sulfide crystalline phases, making them particularly suitable for constructing high-efficiency luminescent processes. For rare earth ion luminescence, the introduction of a low phonon energy environment is of special significance, as it helps alleviate the dissipation of excited state energy through lattice vibration, thereby increasing the probability of luminescent transitions.

From the perspective of material design, the advantage of sulfide glass-ceramics is also reflected in the fact that their crystalline phases can directly undertake the function of high-efficiency luminescence. As a typical sulfide semiconductor microcrystal, CdS has both distinct band luminescence characteristics and tunable local structural environments, and is expected to form a composite system with both transparency and luminescence in a glass matrix. Monisha *et al*. pointed out that the formation of crystalline phases in glass can effectively enhance luminescence intensity and improve the corresponding lifetime characteristics [3]. This rule provides a methodological basis for the introduction of sulfide microcrystals such as CdS: by controlling the crystallization process, a more efficient radiative recombination channel can be established by using crystalline phases while maintaining the processability of glassy materials. Therefore, sulfide glass-ceramics can not only exert the engineering advantages of glass materials, such as easy preparation, formability and suitability for mass processing, but also obtain excellent luminescent responses by virtue of microcrystalline phases such as CdS.

At the application level, glass-ceramics have shown multi-directional potential ranging from near-infrared lighting to optical storage, lasers and temperature sensing. Ji *et al*. suggested that transparent glass-ceramics can be directly used as stable luminescent components in solid-state lighting devices, and exhibit better chemical stability than traditional resin packaging schemes [4]. This trend indicates that the effective coupling of low phonon energy sulfide crystalline phases with glass matrices will endow sulfide glass-ceramics with unique advantages in high-efficiency luminescence, broadband regulation and device stability. Therefore, the systematic study of their preparation fundamentals, luminescent mechanisms, doping regulation and application expansion is of great scientific significance and material application value.

2. Basic Theories and Classification of Sulfide Glass-Ceramics

2.1. Correlation Basis between Structural Characteristics and Luminescent Properties

The luminescent behavior of sulfide glass-ceramics is based on a biphasic structure composed of the glass phase and the crystalline phase in a synergistic manner. The glass phase usually undertakes the role of a continuous network skeleton and component buffering, and its structural integrity, bonding mode and local disorder degree determine the absorption, transfer and non-radiative relaxation background of the matrix for excitation energy. In the discussion of sulfide systems, GeS_2 and Ga_2S_3 are often regarded as the main network units, and the introduction of components such as SiO_2 can be used to adjust the network connection state and local rigidity. Monisha *et al*. found that the crystallization of SiO_2 is accompanied by a change in the number of bridging structures, which further affects the luminescence intensity and lifetime [3]. This insight indicates that the glass phase does not only provide mechanical support, but its network vibration characteristics and defect chemistry are directly involved in the radiative and non-radiative competition process of luminescent centers.

Corresponding to the continuous glass network, the crystalline phase is the key structural unit determining the spectral morphology and high-efficiency luminescence. In transparent glass-ceramics, nanocrystals are usually embedded in the matrix in a dispersed state with particle sizes controlled in the range of several nanometers to tens of nanometers to balance low scattering and effective crystal field construction. Ji *et al*. pointed out that 10–15 nm nanocrystals can maintain the transparency of the material while providing a clear crystal coordination environment for activator ions [4]. Bour *et al*. found that approximately 10 nm crystal grains can stably exist in glass composites and are associated with near-infrared broadband luminescence [7]. For sulfide crystalline phases represented by CdS nanocrystals, the size range of 5–50 nm is of particular structural significance: a smaller size leads to a higher interface ratio and enhanced quantum confinement, making the emission peak position and bandwidth more sensitive to particle size; an increased size improves the internal order of the crystal, but excessively large particles may cause enhanced scattering and local stress accumulation, thereby weakening the effective light output.

The influence of crystallite size on luminescent properties is not limited to the geometric scale itself, but is more reflected in the remodeling of energy level structure and carrier behavior. Nanoscale crystallites change the spatial confinement degree of electron-hole pairs, resulting in changes in the absorption edge, exciton recombination probability and the participation path of defect states. He *et al*. found that microstructure adjustment can cause significant changes in luminescent color and intensity, the root cause of which is related to the change in the relaxation process between ions [6]. In sulfide glass-ceramics with dispersed CdS nanocrystals, a decrease in particle size usually means a stronger quantum size effect, which in turn changes the competitive relationship between intrinsic luminescence and

the luminescence of doping centers; a narrower particle size distribution leads to a more concentrated local potential well distribution, and the luminescence peak shape and color purity are usually more stable.

The glass-crystallite interface is the most active region in the biphasic structure. Due to thermal expansion mismatch, local component segregation and structural rearrangement during crystallization, the interface is often enriched with vacancies, interstitial atoms and coordination-unsaturated sites. On the one hand, these defects may form trap states, inducing the non-radiative dissipation of excitation energy or generating broadband defect luminescence; on the other hand, they may act as energy relays to promote the migration of absorption energy from the glass phase to the luminescent centers of the crystalline phase. Zi *et al*. proposed that the formation of color centers in nanoparticles is an important reason for modulating luminescent behavior, indicating that interface and near-interface defects can significantly change the photoresponse process [1]. If the concentration of interface defects is moderate, an energy transfer channel from the disordered network to the ordered nanocrystals can be formed; if there are too many defects, they will become quenching centers, reducing the quantum efficiency and shortening the lifetime.

Crystalline phase purity is also an important parameter linking structure and luminescence. A high-purity crystalline phase means a more consistent local symmetry, less heterophase interference and a more stable energy level distribution, which is conducive to improving luminescence intensity and reducing inhomogeneous broadening of spectral lines. Liang (2024) *et al*. found that after the precipitation of low phonon energy crystalline phases, both luminescence intensity and lifetime can be enhanced [2]. This result shows that the structural purity of the crystalline phase is not only related to the crystal field environment, but also affects the probability of non-radiative multi-phonon relaxation. For sulfide glass-ceramics, if the CdS crystalline phase is mixed with other ineffective phases or has severe lattice distortion, it is easy to introduce additional scattering and defect recombination paths, damaging both intrinsic luminescence and doped luminescence simultaneously.

The distribution of doping sites determines the upper limit of luminescence efficiency in glass-ceramics. Activator ions can be located in the glass phase, crystalline phase or near the interface, and different sites mean different coordination fields, phonon environments and ion spacings. Ji *et al*. found that after doping ions enter specific coordination centers of nanocrystals, a clear broadband near-infrared emission can be formed [4]. He *et al*. pointed out that element distribution and lifetime measurement can reflect the local aggregation of doping ions and their cross-relaxation changes [6]. This indicates that if rare earth or transition metal ions preferentially enter the low phonon energy, coordination-ordered crystalline phase, their radiative transitions are more likely to be maintained; if they mainly remain in the disordered glass phase or high-defect interface, concentration quenching and non-radiative loss are more likely to occur. Therefore, the core of luminescence optimization is not simply to increase the doping amount, but to regulate the selective distribution of doping ions in the biphasic structure.

Thus, the structural basis of sulfide glass-ceramics can be summarized as follows: the glass network provides a tunable chemical environment and nucleation space, nanocrystals such as CdS endow discrete and high-efficiency luminescent units, and the interface determines whether energy transfer is enhanced or dissipated. The three parameters of size, purity and site distribution are coupled with each other, jointly determining the evolution direction of luminescence peak position, intensity, lifetime and quantum efficiency.

2.2. Classification of Sulfide Glass-Ceramics

Sulfide glass-ceramics can be classified from the dimensions of crystalline phase composition, type of luminescent centers and excitation mode, and different classification frameworks correspond to their structural basis, luminescent mechanisms and application paths respectively. The significance of classification research lies not only in establishing a comparison coordinate between material systems, but also in revealing the coupling relationship between "glass matrix - crystalline phase - luminescent center - excitation condition". Existing research on the luminescence of glass-ceramics shows that the precipitated nano or microcrystalline phases can significantly change the luminescent behavior by reducing the local phonon energy, improving the coordination environment of active ions, regulating the energy transfer channel and maintaining a certain transmittance. Liang *et al*. found that the precipitation of low phonon energy crystalline phases can simultaneously enhance the luminescence intensity and lifetime of rare earth ions, and further support the realization of temperature sensing functions [2]. Monisha *et al*. pointed out that the crystallization process will reconstruct the glass network and improve the radiative performance of luminescent ions, thus making glass-ceramics exhibit better comprehensive chromaticity characteristics in the lighting direction [3]. Such insights provide a methodological basis for the classification of sulfide glass-ceramics.

In terms of the type of crystalline phases, CdS-based sulfide glass-ceramics are the most mature research system and the most representative in luminescence regulation. This type usually takes CdS crystallites as the core luminescent or sensitizing unit, and its advantage is that the crystallite size, crystalline phase distribution and quantum confinement effect can all be used to adjust the absorption edge and emission band, thus having a wide tunable range in the visible to near-infrared region. After CdS crystallites are embedded in glass, they can rely on their own band-edge recombination or surface state participation in luminescence on the one hand, and act as an energy absorption and transfer platform on the other hand, forming a composite luminescent structure with rare earth ions, transition metal ions or other semiconductor components. Existing research on other types of luminescent glass-ceramics shows that there is a significant correlation between nanocrystal size and transparency, and the precipitation of small-scale crystals is conducive to achieving effective luminescence while maintaining high transmittance. Ji *et al*. found that when the size of luminescent nanocrystals is controlled at the level of 10–15 nm and uniformly embedded in glass, the material can simultaneously maintain chemical stability, transparency and near-infrared broadband luminescence capability [4]. This rule has a reference significance for understanding why CdS-based sulfide glass-ceramics have become the dominant system, that is, their research value comes not only from the intrinsically excellent optical response of CdS, but also from its matching with the structural design of transparent glass-ceramics.

ZnS-based sulfide glass-ceramics are usually regarded as an

important supplement to CdS-based systems. Compared with CdS, ZnS-related systems have differences in composition regulation, band structure and luminescence band, and their luminescence can be jointly contributed by intrinsic defects, doping ions and composite quantum states. The classification value of ZnS glass-ceramics is mainly reflected in their ability to serve as a wide band gap sulfide platform for hosting rare earth or transition metal centers, and adjusting the emission color and lifetime through the local crystal field. Bour *et al*. found that different crystallization routes will affect the structural environment of luminescent centers in glass-crystal composites, and further change the near-infrared broadband luminescent behavior [7]. Although this conclusion is from a non-sulfide system, it illustrates that there is a strong correlation between "crystalline phase type - formation mode - luminescent response", and also suggests that the classification of ZnS-based sulfide glass-ceramics cannot only be based on the chemical formula itself, but also be analyzed in combination with its crystallization path and local structural state.

PbS-based sulfide glass-ceramics have a special position in near-infrared luminescence and quantum confinement research. Such systems are usually characterized by a narrow band gap and obvious size-dependent luminescence, and are suitable for discussions oriented to infrared detection, communication bands and broadband near-infrared radiation. The root of their classification independence lies in the fact that PbS crystallites can obtain a strong near-infrared response through particle size adjustment, which is different from the dominant systems in the visible or near-visible region such as CdS and ZnS. However, the toxicity of Pb elements and potential environmental burdens constitute important restrictions on the development of such materials. Therefore, although PbS-based sulfide glass-ceramics are irreplaceable in function, they are often regarded as a representative branch with "outstanding performance but limited application" in classification. Ji *et al*. argued that replacing resin-packaged phosphors with glass-ceramics can improve the chemical stability of devices and reduce the problem of efficiency attenuation [4]. This device-level judgment indicates that if the PbS-based system is used for near-infrared light sources, its classification discussion must simultaneously incorporate the dimensions of stability and safety, rather than taking luminescent performance as the only standard.

Multicomponent sulfide solid solution-based glass-ceramics reflect stronger compositional engineering characteristics. Such materials often realize the synergistic changes of band gap, lattice constant, defect concentration and local crystal field through cation substitution, solid solution regulation or composite crystallization. Therefore, compared with single CdS, ZnS or PbS systems, they have a higher degree of freedom in luminescence design. The classification significance of multicomponent solid solutions lies in that they are no longer limited to the framework of "single crystalline phase determining performance", but emphasize the continuous tunability of luminescence caused by the continuous change of components. He (2024) *et al*. found that the luminescent color can change systematically with the change of doping concentration, the essence of which is closely related to the change of cross-relaxation between active ions [6]. Although this work focuses on the upconversion system, the revealed logic of "component regulation - energy migration - luminescent color evolution" is also applicable to multicomponent sulfide solid solution glass-ceramics, that is, the classification of such materials should highlight their continuous regulation attributes rather than static naming attributes.

Classified by luminescence type, rare earth ion-doped luminescence is the core direction in sulfide glass-ceramics. The core reason is that rare earth ions have a relatively stable 4f electronic configuration, a rich energy level structure and diverse upconversion, downconversion and near-infrared emission channels; while sulfide crystalline phases usually have low phonon energy, which can inhibit multi-phonon non-radiative relaxation, thereby improving the luminescence efficiency of rare earth ions. Liang *et al*. found that the precipitation of low phonon energy crystalline phases helps enhance the upconversion luminescence and lifetime of Er^{3+} ions [2]. Zhang *et al*. pointed out that the selection of crystalline phase hosts directly affects the upconversion luminescence characteristics, and high-efficiency blue light output can be achieved by adjusting the Yb^{3+} concentration, but excessive doping will cause concentration quenching [5]. Zi *et al*. proposed that rare earth co-doped glass-ceramics can couple reversible photochromic behavior with upconversion luminescence modulation, thus showing repeatable modulation functions for optical storage [1]. These studies jointly show that in sulfide glass-ceramics, rare earth ion-doped luminescence constitutes the main axis in the classification not only because of its rich luminescent spectral lines, but also because it is highly sensitive to local structure, energy transfer and external field response, suitable for fine regulation.

Intrinsic defect luminescence is another type of luminescence that cannot be ignored. Especially in sulfide glass-ceramics, sulfur vacancies, metal vacancies, interstitial atoms and interface defects may all form energy level centers and participate in absorption, capture and recombination processes. Such luminescence generally shows broadband characteristics and is closely related to the material preparation atmosphere, heat treatment system, grain size and glass network rearrangement. Zi *et al*. found that the formation of color centers in nanocrystals is an important driving factor for material color development and luminescence modulation [1]. This insight indicates that defect centers are not simply "by-products", but may become key units determining optical functions. Therefore, in the classification of sulfide glass-ceramics, it is necessary to propose defect luminescence independently, especially when explaining broadband luminescence, afterglow, photochromic coupling and nonlinear photoresponse.

Transition metal-doped luminescence reflects another typical mechanism, which is characterized by the fact that d-electron transitions are significantly affected by the crystal field environment, so the luminescence peak position, bandwidth and lifetime are extremely sensitive to the local structure of crystallites. Bour *et al*. found that Ni^{2+}-doped glass-ceramics can achieve broadband near-infrared emission centered at approximately 1300 nm, and the material remains transparent in the near-infrared region [7]. Ji *et al*. pointed out that when Cr^{3+} is located at the octahedral center of nanocrystals, it can produce broadband near-infrared emission covering 600–1400 nm under visible light excitation [4]. These results indicate that transition metal ion-doped luminescence should be listed as an important branch alongside rare earth luminescence in the classification of glass-ceramics, because it usually has a wider emission band, stronger crystal field

dependence and more distinct application orientation, especially suitable for broadband near-infrared light sources, imaging and sensing.

Quantum dot composite luminescence emphasizes the synergistic effect between semiconductor crystallites and other luminescent centers. In sulfide glass-ceramics, quantum dots can be independent luminescent bodies, as well as sensitizers, energy relays or color purity control units. The key of this classification method does not lie in whether a single crystallite luminesces, but in whether it forms a coupled composite structure of "quantum dot - glass matrix - doping ion". Ji *et al*. found that the integrated structure composed of luminescent nanocrystals and glass can support stable device integration and is superior to the traditional powder/resin packaging mode [4]. From this perspective, quantum dot composite luminescence is an important category for sulfide glass-ceramics to develop from basic luminescent materials to integrated light sources and functional devices, and its classification value is closely related to application transformation.

Classified by excitation mode, photoluminescence is the most commonly used and fully studied type in sulfide glass-ceramics. Such systems usually use ultraviolet, visible or near-infrared light as the excitation source, and obtain emission output through interband absorption, energy transfer, upconversion absorption or defect excitation. Most studies on luminescence enhancement, luminescent color tuning and lifetime extension are based on the photoluminescence framework. He (2024) *et al*. found that adjusting the Er^{3+} doping concentration can significantly change the upconversion luminescence intensity and color, and cross-relaxation is the main reason for the transition from green to red [6]. Zhang *et al*. pointed out that adjusting the Yb^{3+} content under 980 nm pumping can achieve high-efficiency blue upconversion luminescence of Tm^{3+} [5]. Monisha *et al*. found that after crystallization induced by heat treatment, the material exhibits enhanced luminescence under different photoexcitation conditions [3]. These works show that the classification of photoluminescence is not only a classification in experimental means, but also directly corresponds to the differences in the population mode of excited states and energy flow paths in sulfide glass-ceramics, so it should be placed at the center of the classification system.

The electroluminescence classification is more device-oriented, and its core concern is the luminescence ability of materials under electric field, carrier injection or composite conditions. For sulfide glass-ceramics, the significance of this classification is to expand them from optical functional materials to candidates for active luminescent devices. Although photoluminescence is more commonly discussed in existing glass-ceramic research, from the characteristic that the integrated glass-ceramic structure can balance transparency, stability and formability, the electroluminescence system has independent classification value. Ji (2025) *et al*. argued that transparent luminescent glass-ceramics can directly construct stable solid-state lighting devices [4]. Although this result is manifested as a light-conversion LED, it shows that the integrability of glass-ceramics at the device level is relatively clear. Therefore, the electroluminescence classification can be regarded as an extension direction of sulfide glass-ceramics for display, lighting and on-chip light sources.

Radioluminescence belongs to a more application-oriented classification, focusing on the photoresponse under high-energy particles, X-rays or other irradiation conditions. For sulfide glass-ceramics, the importance of this classification is that sulfide crystallites often have high light absorption capacity and fast radiation response potential, suitable for development in the directions of detection, imaging and safety monitoring. Although existing relevant glass-ceramic research mostly focuses on photoexcitation, results such as reversible photoresponse, near-infrared broadband emission and stable device construction have shown the possibility of glass-ceramic systems to achieve functional output under complex excitation environments. Zi *et al*. found that the reversible optical modulation in glass-ceramics has excellent repeatability and anti-fatigue characteristics [1]. This conclusion suggests that if sulfide glass-ceramics are extended to radioluminescence scenarios, their structural stability and cycle reliability will become important considerations in the classification discussion.

From the perspective of classification logic, the type of crystalline phase determines the band basis and local structural framework of sulfide glass-ceramics, the luminescence type determines the dominant luminescent center and radiation mechanism, and the excitation mode determines the energy-carrying input path and application scenario. The three classification methods are not independent of each other, but cross-constitute the basic coordinate system for the research of sulfide glass-ceramics. Taking the CdS-based rare earth-doped photoluminescence system as an example, its classification label includes three meanings: "CdS crystalline phase", "rare earth ion luminescence" and "photoexcitation"; if quantum dot composite or transition metal co-doping is further introduced, more complex subcategories will be formed. It can be seen that the classification of sulfide glass-ceramics should not stay at simple naming, but should be carried out around the coupling relationship between the crystalline phase, luminescent center and excitation process, which is also the theoretical premise for the subsequent discussion of their preparation, luminescent mechanisms, doping regulation and application progress.

2.3. Core Evaluation Indices of Luminescent Properties

The evaluation of luminescent properties needs to be based on quantifiable spectral parameters, among which emission intensity, quantum yield, fluorescence lifetime, emission peak position and full width at half maximum, chromaticity parameters, as well as thermal stability and modulation stability constitute the core indices of the glass-ceramic system. For sulfide glass-ceramics, these indices not only reflect the intrinsic radiation capacity of luminescent centers, but also characterize the coupling relationship between crystallite precipitation, local crystal field, phonon energy and doping concentration.

Emission intensity is the most direct characterization parameter, usually obtained by steady-state excitation-emission spectroscopy, comparing the relative or absolute intensity of characteristic peaks under unit excitation conditions. Its value is jointly controlled by the content of luminescent centers, absorption efficiency, energy transfer efficiency and non-radiative relaxation probability. Liang *et al*. found that after the precipitation of low phonon energy crystalline phases, upconversion luminescence and lifetime are enhanced synchronously, indicating that the crystalline phase can effectively inhibit non-radiative loss and improve

the probability of radiative recombination [2]. He *et al*. found that after adjusting the Er^{3+} concentration, the upconversion intensity can change by more than 100 times, and the luminescent color changes from green to red at the same time, which indicates that the intensity evaluation cannot be discussed separately from the cross-relaxation process between energy levels [6]. Extended to CdS-based sulfide glass-ceramics, the emission intensity is usually also significantly affected by crystallite size, distribution uniformity and interface defect density, because these factors determine the competitive relationship between exciton recombination and trap capture.

Quantum yield is used to characterize the efficiency of converting absorbed photons into emitted photons, which can be divided into external quantum yield and internal quantum yield, and absolute measurement is often realized with the help of an integrating sphere in testing. This index can comprehensively reflect the balance between material absorption, radiative transition and non-radiative attenuation. Ji *et al*. found that the internal quantum efficiency of Cr^{3+}-doped transparent glass-ceramics under blue light excitation is about 0.81%, and thus proved that transparent glass-ceramics can be used as a stable near-infrared luminescent conversion medium [4]. For rare earth-doped sulfide glass-ceramics, quantum yield is particularly important because the f-f transition of rare earth ions is relatively sensitive, and the host low phonon energy environment, hydroxyl content and crystalline phase enrichment degree all affect the quantum efficiency; in CdS-based systems, quantum yield is more often used to judge whether the quantum confinement effect and surface defect passivation are effective.

Fluorescence lifetime reflects the average residence time of excited state particles, usually obtained by fitting the time-resolved decay curve under pulse excitation. A longer lifetime generally means weaker non-radiative processes, but its interpretation still needs to be comprehensively analyzed in combination with energy migration and concentration quenching. Monisha *et al*. (2024) found that after the formation of crystalline phases by heat treatment, the decay lifetime of Dy^{3+} and Sm^{3+} is increased to the microsecond level, indicating that the precipitation of crystallites improves the local luminescent environment of rare earth ions [3]. Liang *et al*. pointed out that crystallization not only enhances the Er^{3+} upconversion emission intensity, but also prolongs its lifetime, indicating that lifetime measurement is an important basis for judging whether structural optimization in glass-ceramics is effective [2]. In sulfide glass-ceramics, the lifetime of rare earth-doped systems can also be used to distinguish whether ions enter the glass phase or the crystalline phase; the CdS-based system can use time-resolved results to judge the relative contribution of band-edge emission and defect emission.

Emission peak position and full width at half maximum are used to describe the position of the luminescence band and the spectral coverage range, obtained by the peak value of the emission spectrum and peak shape fitting respectively. The change of peak position often corresponds to the change of crystal field strength, particle size effect or energy level splitting state; the full width at half maximum is related to broadband lighting and spectral matching ability. Ji *et al*. found that the near-infrared emission peak position of Cr^{3+}-doped glass-ceramics is about 810 nm with a full width at half maximum of about 230 nm, showing broadband luminescence characteristics, suitable for night vision lighting conversion applications [4]. Bour *et al*. found that Ni^{2+}-doped glass-ceramics exhibit broadband emission at approximately 1300 nm with a full width at half maximum of 228 nm, indicating that transition metal luminescent centers can achieve relatively wide spectral line output in glass-ceramics [7]. For CdS-based sulfide glass-ceramics, the evaluation of peak position and bandwidth is particularly critical because they often directly correspond to the crystallite size distribution and the degree of quantum confinement.

Chromaticity parameters are an important part of the application evaluation of visible luminescence systems, usually obtained by converting the emission spectrum into CIE coordinates, correlated color temperature or color purity. He *et al*. found that the change in Er^{3+} concentration will cause a significant change in upconversion color with higher color purity, indicating that chromaticity indices can sensitively reflect the effect of doping regulation [6]. Zhang *et al*. pointed out that Yb^{3+}/Tm^{3+} co-doped samples exhibit chromatic coordinates in the blue region under optimal luminescence conditions, and excessive Yb^{3+} concentration will cause concentration quenching, indicating that chromatic coordinates should be investigated synchronously with intensity optimization [5].

Stability indices are oriented to practical applications, including thermal repeatability, cycle modulation capability and anti-fatigue property. Zi *et al*. found that the maximum luminescence modulation rate can reach 79.83%, and the cycle process has good repeatability and anti-fatigue property, indicating that dynamic response capability has become an important dimension for the evaluation of functional glass-ceramics [1]. Therefore, the core evaluation of the luminescent properties of sulfide glass-ceramics should not stay at a single brightness comparison, but take quantum yield and lifetime as efficiency indices, peak position, bandwidth and chromaticity as spectral indices, and stability as application indices, so as to accurately reveal the structure-luminescence correlation of CdS-based and rare earth-doped systems.

3. Luminescent Mechanisms and Doping Regulation Mechanisms of Sulfide Glass-Ceramics

3.1. Intrinsic Luminescent Mechanisms

Intrinsic luminescent mechanisms mainly depend on the internal band structure of sulfide crystallites, quantum confinement effect and the local energy level distribution induced by intrinsic point defects. For sulfide glass-ceramic systems represented by CdS crystallites, their luminescence can usually be classified into two types: exciton-related luminescence and defect-related luminescence; the two are not isolated from each other, but show luminescent behavior with both competition and synergy under the combined action of crystallite size, crystalline phase purity and defect concentration. The glass-crystallite composite structure restricts the grain growth in space and provides a non-equilibrium environment for the formation and stabilization of defects at the same time, so the intrinsic luminescence is particularly sensitive to the preparation conditions.

The exciton luminescence of CdS crystallites essentially originates from the band-edge radiative transition corresponding to the recombination of electrons and holes. When the grain size is reduced to the nanometer scale, the movement of carriers is restricted by space, and the positions of the conduction band bottom and valence band top shift

with the change of particle size, resulting in an increase in the effective band gap and a blue shift of the emission peak. This rule can usually be described by the Brus formula, that is, the crystallite band gap not only includes the intrinsic band gap term of bulk semiconductors, but also superimposes the quantum confinement term inversely proportional to the square of the particle size and the correction term of electron-hole Coulomb interaction. It can be seen that the smaller the grain size, the stronger the confinement effect and the shorter the emission wavelength; as the grain size increases, the band gap gradually approaches that of bulk CdS, and the emission shifts back to the long-wave direction. The nanocrystalline scale in transparent glass-ceramics has a decisive effect on the luminescent behavior. Ji *et al*. found that when the nanocrystal size is at the level of 10–15 nm, the crystallites can be stably precipitated in the glass matrix and exhibit obvious crystal field-related luminescence. This result shows that the precipitation of nanoscale crystalline phases can effectively change the local energy level structure and establish a direct relationship between size and luminescent response [4].

In sulfide glass-ceramics, the exciton luminescence intensity is not only determined by the particle size alone, but also closely related to the degree of crystallite precipitation and crystallization state. Moderate heat treatment can promote the uniform precipitation of crystallites, improve the local order, thereby reducing the probability of non-radiative relaxation and making band-edge radiative recombination more likely to occur; if the heat treatment is insufficient, the crystallization is incomplete and the volume fraction of crystallites is low, making it difficult to form stable exciton luminescent centers; if the heat treatment is excessive, the grains coarsen, scattering is enhanced, and even new non-radiative recombination channels may be induced, weakening the luminescence output. Liang *et al*. found that there is an obvious optimal window for crystallization conditions, and a moderate heat treatment temperature and time can make the crystalline phase promote luminescence enhancement, while deviating from this window is not conducive to the improvement of luminescent performance [2]. This insight also has methodological significance for CdS glass-ceramics, that is, regulating the grain size distribution and crystallization degree by controlling the heat treatment system is the key way to realize the synergistic optimization of exciton emission wavelength and intensity.

In addition to band-edge exciton luminescence, defect luminescence often occupies an important position in sulfide glass-ceramics. Due to the relatively low bond energy of sulfides, anion or cation vacancies are easy to form during melting, quenching and subsequent heat treatment processes, among which sulfur vacancies (V_S) and cadmium vacancies (V_Cd) are the two most representative types of intrinsic defects in the CdS system. V_S can usually be formed under conditions of sulfur deficiency, local reduction or high-temperature volatilization, and its corresponding defect energy level is generally closer to the conduction band. After electrons are captured by defects and recombine with valence band holes, high-energy visible emission can be generated, often showing green light; V_Cd is more likely to be formed when cadmium is insufficient, local oxidation or the composition deviates from the stoichiometric ratio, and its acceptor-type defect energy level is relatively deeper, and the energy released by radiative recombination is lower, thus often corresponding to red light emission. The relative abundance of the two types of defect centers determines the broadband luminescence morphology and comprehensive chromatic coordinates of the sample.

The formation mechanism of defect luminescence can be understood as follows: the mismatch of the local coordination environment during the preparation process introduces trap states of different depths. Electrons and holes generated by photoexcitation are captured by these traps during the relaxation process, and then release photons through defect-assisted recombination. If the V_S concentration is high, the green light-related defect emission is dominant; if the proportion of V_Cd increases, the red light component is enhanced. When the exciton state is close to the defect state in energy, excitons may also transfer to the defect state, thereby weakening the band-edge luminescence and enhancing the broadband defect emission. Therefore, the intrinsic luminescence in sulfide glass-ceramics often shows the characteristics of "size tuning the band edge, defects determining the hue": the particle size mainly controls the basic framework of the band gap and emission position, and defects further shape the emission band width, peak shape and comprehensive color performance.

There is a significant coupling relationship between defect concentration and preparation process. The glass melting atmosphere, raw material stoichiometric ratio, cooling rate and heat treatment temperature all affect the vacancy formation energy and defect freezing degree. Conditions with strong reducibility or serious sulfur loss are more likely to increase the V_S concentration, thereby enhancing the green light defect emission; on the contrary, when the cadmium sites are more prone to mismatch or migration, the red light emission related to V_Cd will be more obvious. Zi *et al*. found that the formation of color centers in transparent glass-ceramics is closely related to their reversible optical response, indicating that heat treatment and atmosphere control can effectively regulate the generation and stability of defect centers [1]. Although this result is from another type of glass-ceramic system, the revealed correlation of "process - defect - luminescence" has a direct enlightening significance for understanding the vacancy regulation in CdS glass-ceramics, that is, intrinsic defects are not inevitable by-products, but structural units that can be directionally regulated through process design.

Heat treatment temperature is the core parameter for regulating the purity and defect concentration of the CdS crystalline phase. On the one hand, appropriately increasing the heat treatment temperature is conducive to the precipitation of CdS crystallites from the disordered glass matrix and the improvement of crystal structure, increasing the purity of the crystalline phase and reducing the non-radiative loss caused by structural disorder; on the other hand, an excessively high temperature may aggravate sulfur volatilization, induce vacancy enrichment, cause abnormal enhancement of defect luminescence and even luminescence quenching. Monisha *et al*. found that heat treatment will change the band-edge position and luminescence intensity in glass-ceramics, and reasonable heat treatment can enhance luminescence [3]. It can be inferred that in CdS glass-ceramics, heat treatment not only affects the grain size and its distribution, but also determines the relative proportion of exciton luminescence and defect luminescence by changing the local chemical environment and defect formation behavior. Only by striking a balance between crystalline phase precipitation and defect inhibition can a higher intrinsic

luminescence efficiency be obtained.

From the perspective of luminescence enhancement mechanism, the improvement of crystalline phase purity can reduce the scattering and non-radiative recombination caused by heterophase interfaces and disordered regions, making the exciton radiation process more effective; a moderate reduction in defect concentration can weaken deep trap capture and improve the probability of direct carrier recombination; if the goal is to obtain a specific hue of broadband intrinsic emission, a certain proportion of V_S or V_Cd can also be retained by controlled defects to achieve the tunable output of green or red light components. Therefore, the optimization of intrinsic luminescence is not simply the pursuit of "the fewer defects the better", but the fine balance of defect types and concentrations on the premise of ensuring the purity of the crystalline phase and the smoothness of the radiation channel. He *et al*. found that the luminescent color will change systematically with the change of interionic interaction and local structure after crystallite precipitation, indicating that the adjustment of the crystallite environment can significantly change the competitive relationship between emission channels [6]. Mapping this conclusion to the CdS intrinsic luminescence system, it can be understood that the internal structural ordering of crystallites and the reconstruction of local defect states jointly determine the final luminescence spectrum shape.

In general, the intrinsic luminescence of CdS sulfide glass-ceramics follows a clear structure-energy level-spectral correlation: the nanocrystal size adjusts the band gap and controls the exciton emission wavelength through quantum confinement, intrinsic vacancies such as V_S and V_Cd endow green and red light defect emission components respectively, and process parameters such as heat treatment temperature further determine the intrinsic luminescence intensity and spectral shape distribution by affecting the crystalline phase purity, grain size and defect formation behavior. The main path to enhance the band-edge intrinsic luminescence intensity is to realize the precipitation of high-purity CdS crystallites and inhibit excessive defects through process optimization; the controlled retention of specific defect centers can expand the regulation space of broadband luminescence and comprehensive color. This internal law constitutes an important theoretical basis for subsequent doping regulation and device application design.

3.2. Doped Luminescence Mechanism

The doped luminescence mechanism is the core of realizing spectrally designable output for sulfide glass-ceramics. Its essence lies in the competitive relationship among energy level splitting, radiative transition and non-radiative relaxation of doped ions in the composite local structure of glass phase and crystallite phase, as well as the energy migration process between sensitizers and activators. After the precipitation of crystallites, the luminescent centers can enrich from the disordered glass environment to the crystallite sites with more definite coordination and more stable crystal field, thereby changing the transition probability, spectral band morphology, luminescence lifetime and thermal stability. Relevant studies on glass-ceramics have shown that the formation of low phonon energy crystallite phases can usually inhibit multi-phonon relaxation and improve the luminescence efficiency of rare earth ions, and thus this rule has important reference significance for sulfide glass-ceramics that also emphasize a low phonon energy environment.

The exciton luminescence of CdS crystallites essentially originates from the band-edge radiative transitions corresponding to the recombination of electrons and holes. When the grain size is reduced to the nanometer scale, the motion of carriers is spatially confined, and the positions of the conduction band bottom and valence band top shift with the variation of particle size, leading to an increase in the effective band gap and a blue shift of the emission peak. This rule can usually be described by the Brus equation: the band gap of crystallites includes not only the intrinsic band gap term of bulk semiconductors, but also a superposed quantum confinement term inversely proportional to the square of the particle size and a correction term for electron-hole Coulomb interaction. It is thus evident that the smaller the grain size, the stronger the confinement effect and the shorter the emission wavelength; as the grain size increases, the band gap gradually approaches that of bulk CdS, and the emission shifts back to the long-wave direction. The size of nanocrystals exerts a decisive effect on the luminescent behavior of transparent glass-ceramics. Ji *et al*. proposed that when the nanocrystal size is in the range of 10–15 nm, the crystallites can be stably precipitated in the glass matrix and exhibit distinct crystal field-related luminescence. This result indicates that the precipitation of nanoscale crystalline phases can effectively modify the local energy level structure and establish a direct correlation between size and luminescent response [4].

In sulfide glass-ceramics, the intensity of exciton luminescence is not determined by particle size alone, but is also closely related to the degree of crystallite precipitation and crystallization state. Moderate heat treatment can promote the uniform precipitation of crystallites, improve the local orderliness, thereby reducing the probability of non-radiative relaxation and facilitating the occurrence of band-edge radiative recombination. Insufficient heat treatment will lead to incomplete crystallization and a low volume fraction of crystallites, making it difficult to form stable exciton luminescent centers. Excessive heat treatment, on the other hand, will cause grain coarsening and enhanced scattering, and may even induce new non-radiative recombination channels that weaken the luminescence output. Liang *et al*. suggested that there exists a distinct optimal window for crystallization conditions; a moderate heat treatment temperature and duration can enable the crystallized phase to enhance luminescence, while deviating from this window is detrimental to the improvement of luminescent performance [2]. This insight also has methodological significance for CdS glass-ceramics, namely that regulating the grain size distribution and the degree of crystallization by controlling the heat treatment regime is the key approach to achieving the synergistic optimization of the wavelength and intensity of exciton emission.

In addition to band-edge exciton luminescence, defect luminescence often plays an important role in sulfide glass-ceramics. Due to the relatively low bond energy of sulfides, anionic or cationic vacancies are prone to form during melting, quenching and subsequent heat treatment processes, among which sulfur vacancies (V_S) and cadmium vacancies (V_Cd) are the two most representative intrinsic defects in the CdS system. V_S can usually form under conditions of sulfur deficiency, local reduction or high-temperature volatilization, and its corresponding defect energy level is generally closer to the conduction band. After electrons are

captured by the defects and recombine with valence band holes, high-energy visible emission can be generated, which is typically manifested as green light. In contrast, V_Cd is more likely to form when cadmium is insufficient, under local oxidation or when the composition deviates from the stoichiometric ratio; its acceptor-type defect energy level is relatively deeper, and the energy released by radiative recombination is lower, thus it usually corresponds to red light emission. The relative abundance of these two types of defect centers determines the broadband luminescence morphology and the comprehensive chromatic coordinates of the sample.

The formation mechanism of defect luminescence can be understood as follows: mismatches in the local coordination environment during the preparation process introduce trap states of varying depths; electrons and holes generated by photoexcitation are captured by these traps during relaxation, and photons are subsequently released through defect-assisted recombination. When the concentration of V_S is high, green light-related defect emission dominates; when the proportion of V_Cd rises, the red-light component is enhanced. When the exciton state is energetically close to the defect state, excitons may also transfer to the defect state, thereby weakening band-edge luminescence and enhancing broadband defect emission. Therefore, intrinsic luminescence in sulfide glass-ceramics typically exhibits the characteristic of "particle size tuning the band edge, defects determining the hue": the particle size mainly controls the basic framework of the band gap and emission position, while defects further shape the emission band width, peak profile and comprehensive color performance.

There is a significant coupling relationship between defect concentration and preparation processes. The glass melting atmosphere, raw material stoichiometric ratio, cooling rate and heat treatment temperature all affect the vacancy formation energy and the degree of defect freezing. Conditions with strong reducibility or severe sulfur loss are more likely to increase the concentration of V_S, thereby enhancing green light defect emission; on the contrary, when cadmium sites are more prone to mismatch or migration, the red-light emission related to V_Cd will become more pronounced. Zi *et al*. proposed that the formation of color centers in transparent glass-ceramics is closely correlated with their reversible optical response, indicating that heat treatment and atmosphere control can effectively regulate the generation and stability of defect centers [1]. Although this result is derived from another type of glass-ceramic system, the revealed "process-defect-luminescence" correlation provides direct enlightenment for understanding vacancy regulation in CdS glass-ceramics, namely that intrinsic defects are not inevitable by-products but structural units that can be directionally regulated through process design.

Heat treatment temperature is the core parameter for regulating the crystalline phase purity and defect concentration of CdS. On the one hand, a moderate increase in heat treatment temperature facilitates the precipitation of CdS crystallites from the disordered glass matrix and the perfection of crystal structure, which improves the crystalline phase purity and reduces the non-radiative losses caused by structural disorder. On the other hand, an excessively high temperature may aggravate sulfur volatilization and induce vacancy enrichment, leading to an abnormal enhancement of defect luminescence and even luminescence quenching. Monisha *et al*. pointed out that heat treatment can alter the band edge position and luminescence intensity in glass-ceramics, and rational heat treatment is capable of enhancing luminescence [3]. It can thus be inferred that in CdS glass-ceramics, heat treatment not only affects the grain size and its distribution, but also determines the relative proportion of exciton luminescence and defect luminescence by changing the local chemical environment and defect formation behavior. Only by striking a balance between crystalline phase precipitation and defect inhibition can a higher intrinsic luminescence efficiency be achieved.

From the perspective of luminescence enhancement mechanisms, an increase in crystalline phase purity can reduce scattering and non-radiative recombination caused by heterophase interfaces and disordered regions, thus making the exciton radiation process more efficient; a moderate reduction in defect concentration can weaken deep trap capture and improve the probability of direct carrier recombination; if the goal is to achieve broadband intrinsic emission with a specific hue, a certain proportion of V_S or V Cd can also be retained via controlled defects to realize tunable output of green or red light components. Therefore, the optimization of intrinsic luminescence does not simply pursue the notion of "the fewer defects the better", but rather involves the delicate balancing of defect types and concentrations on the premise of ensuring crystalline phase purity and unobstructed radiation channels. He *et al*. proposed that the luminescent color changes systematically with the variation of interionic interactions and local structures after crystallite precipitation, indicating that the modulation of the crystallite microenvironment can significantly alter the competitive relationship among different emission channels. When this conclusion is applied to the CdS intrinsic luminescence system, it can be understood that the internal structural ordering of crystallites and the reconstruction of local defect states jointly determine the final luminescence spectral profile.

In general, the intrinsic luminescence of CdS sulfide glass-ceramics follows a clear structure-energy level-spectra correlation: the nanocrystal size modulates the band gap and controls the exciton emission wavelength via the quantum confinement effect; intrinsic vacancies such as V_S and V_Cd endow the material with green and red light defect emission components, respectively; and process parameters including heat treatment temperature further determine the intensity and spectral profile of intrinsic luminescence by influencing the crystalline phase purity, grain size and defect formation behavior. Realizing the precipitation of high-purity CdS crystallites and inhibiting excessive defects through process optimization constitutes the primary approach to enhancing the intensity of band-edge intrinsic luminescence; meanwhile, retaining specific defect centers in a controlled manner enables the expansion of the regulatory space for broadband luminescence and comprehensive color performance. This inherent rule forms an important theoretical foundation for subsequent doping regulation and the design of device applications.

3.2 Doped Luminescence Mechanism

The doped luminescence mechanism is the core of enabling spectrally designable emission in sulfide glass-ceramics. Its essence lies in the competitive relationship among energy level splitting, radiative transition and non-radiative relaxation of doped ions within the composite local structure of glass phase and crystallite phase, as well as the energy migration process between sensitizers and activators. After

the precipitation of crystallites, luminescent centers can migrate from the disordered glass matrix to crystallite sites with more defined coordination and more stable crystal fields, thereby altering the transition probability, spectral band morphology, luminescence lifetime and thermal stability. Relevant studies on glass-ceramics have demonstrated that the formation of low phonon energy crystallite phases can typically suppress multi-phonon relaxation and enhance the luminescence efficiency of rare earth ions, and thus this rule holds important reference significance for sulfide glass-ceramics that also emphasize a low phonon energy environment.

Luminescence from rare earth ion doping mainly involves two major types of transitions: intra-4f electron shell transitions and 5d-related transitions. The 4f-4f transitions are shielded by the outer 5s and 5p electrons, yielding emission peaks with narrow linewidths and high color purity, which makes them suitable for generating characteristic red, green and near-infrared luminescence. In sulfide glass-ceramics, when ions such as Eu^{3+} and Tb^{3+} occupy crystallographic sites with low local symmetry, their electric dipole transition probabilities can be enhanced, thus facilitating the $^5D_0 \rightarrow {}^7F_2$ red emission of Eu^{3+} and the $^5D_4 \rightarrow {}^7F_5$ green emission of Tb^{3+}. The effect of microcrystallization is reflected not only in crystal field modulation, but also in the reconstruction of the spatial distribution of luminescent centers: when rare earth ions are preferentially enriched in the microcrystalline regions, the local vibrational energy is reduced, the coordination order is elevated, and radiative recombination becomes dominant. For upconversion luminescent systems, the doping mechanism is more prominently manifested as a coupled process of sensitized absorption, stepwise energy transfer and interionic cross-relaxation. Yb^{3+}, with a large absorption cross-section at 980 nm, is often used as a sensitizer to transfer energy to Er^{3+}, Tm^{3+} or Tb^{3+} for improving the efficiency of visible or near-infrared luminescence under near-infrared excitation. In co-doped systems, Yb^{3+} first absorbs pump photons and is excited to the excited state, then transfers energy to activator ions through an energy matching process, enabling the latter to undergo multi-step excitation and generate upconversion radiation. This result indicates that the doping concentration not only determines the absorption capacity, but also profoundly affects the interionic distance and energy migration pathways. In sulfide glass-ceramics, if Yb^{3+}/Er^{3+} are co-doped in the CdS-based microcrystalline regions, the low phonon energy environment will help maintain the population of intermediate excited states and reduce non-radiative losses, thereby being more conducive to near-infrared upconversion luminescence and its application in fiber laser-related devices.

The efficiency of energy transfer between Yb^{3+} and other activator ions is also closely related to the co-doping ratio. An excessively low content of the sensitizer will limit pump absorption, while an excessively high content may induce energy migration to defect centers and cause concentration quenching. These phenomena indicate that the doped luminescence mechanism in sulfide glass-ceramics cannot be merely understood as the energy level transition of isolated ions, but must be analyzed in combination with the competitive absorption and energy dissipation processes caused by internal defects, color centers and ion aggregation in the crystallites. In addition to narrowband 4f-4f luminescence, the 5d-related transitions of rare earth ions are also of great significance in sulfide glass-ceramics. The 4f-5d or 5d-4f transitions of ions such as Ce^{3+} and Eu^{2+} are more sensitive to the crystal field and covalent environment, thus typically exhibiting broadband emission that can cover the ultraviolet, visible and even near-infrared regions. Since the 5d orbitals are exposed to the external coordination field, the composition of the crystallite phase, anion polarizability and local structural distortion can all cause shifts in the emission peak position and changes in the band width. This insight shows that when discussing the luminescence of Eu^{2+} or Ce^{3+} in sulfide glass-ceramics, the local coordination environment and valence state stability must be incorporated into a unified framework: on the one hand, the high anion polarizability of sulfides is conducive to enhancing the response of 5d energy levels to the crystal field; on the other hand, the structural inhomogeneity at the crystallite/glass interface may lead to the diversification of luminescent centers, thereby forming composite broadband emission.

The Red Luminescence Mechanism of Eu^{3+}-doped Sulfide Glass-ceramics The red luminescence mechanism of Eu^{3+}-doped sulfide glass-ceramics can be attributed to three processes: matrix absorption, energy transfer, and radiative recombination at the characteristic energy levels of Eu^{3+}. If crystallite phases such as ZnS and CdS exhibit strong band-edge absorption or defect-state absorption, they can first act as light-harvesting units. Subsequently, they excite the upper energy levels of Eu^{3+} through non-radiative energy transfer, ultimately generating red light via the $^5D_0 \rightarrow {}^7F\_J$ energy level transition. Among these, the $^5D_0 \rightarrow {}^7F_2$ transition is often regarded as a key emission channel for evaluating the local non-centrosymmetry. After crystallite precipitation, if Eu^{3+} preferentially occupies lattice sites with fewer defects and moderate symmetry, the luminescence intensity can be improved while maintaining spectral resolution; however, if it enriches at the interface or in high-defect regions, quenching may occur due to cross-relaxation and defect capture. Thus, the reason why Eu^{3+}-doped ZnS/CdS composite glass-ceramics are suitable for LED red light conversion lies not only in the stable intrinsic emission of rare earth ions, but also in the fact that semiconductor crystallites can provide additional sensitized absorption channels. ### Quantum Dot/Rare Earth Composite Doping Quantum dot/rare earth composite doping represents a highly representative synergistic luminescence mechanism in sulfide glass-ceramics. CdS quantum dots can serve as sensitizers, efficiently capturing excitation energy by virtue of their large absorption cross-section and tunable exciton energy levels. Rare earth ions, as activators, are responsible for the output of characteristic narrowband or near-infrared radiation. The key issue in this composite system lies in the energy matching between the exciton states, surface defect states of quantum dots, and the 4f or 5d energy levels of rare earth ions. When the emission energy of CdS quantum dots is higher than the acceptor energy levels of rare earth ions, energy can be transferred to the rare earth ions through radiative reabsorption or non-radiative resonance, combining the advantages of the "broad absorption" of quantum dots and the "narrow emission" of rare earth ions. The introduction of rare earth ions may also suppress quantum dot blinking and thermal quenching by capturing energy related to surface defects and altering the interfacial charge distribution. This mechanism implies that composite doping is not a simple superposition of two luminescent centers, but rather establishes a new balance between intensity and stability

through a cascade pathway of "semiconductor absorption—interfacial energy transfer—rare earth emission". ### Transition Metal Ion Doping Transition metal ion doping exhibits luminescence rules that are significantly different from those of rare earth ions. The luminescence of ions such as Mn^{2+} and Cu^{2+} typically originates from d-d transitions. Their energy level positions are highly correlated with the crystal field strength, spin selection rules, and local coordination geometry, resulting in broad emission peak shapes and greater sensitivity to matrix composition, coordination distortion, and defect states. In contrast to the shielded 4f electrons of rare earth ions, which yield relatively stable spectral lines, the d orbitals of transition metal ions are directly exposed to the coordination environment. This makes their luminescence color and efficiency more easily modulated by the type of crystallite phase and local symmetry. It can be inferred that the luminescence of transition metal ions such as Mn^{2+} and Cu^{2+} in sulfide glass-ceramics will also strongly depend on tetrahedral/octahedral coordination selection, crystal field splitting magnitude, and defect compensation methods. Their advantage lies in the easy achievement of broadband tunable emission, while their limitation lies in the usually more significant non-radiative loss and concentration quenching. ### Doping Regulation Perspective From the perspective of doping regulation, the luminescence mechanism is ultimately governed by three types of factors: the type and valence state of doping ions, the local structure provided by the crystallite phase, and the interionic interactions induced by doping concentration. Rare earth ions are more suitable for obtaining high color purity narrowband emission and high-stability near-infrared output. Transition metal ions are more suitable for achieving broadband tunable luminescence. The quantum dot/rare earth composite strategy combines the advantages of broad absorption and characteristic emission. Due to their low phonon energy and high anion polarizability, sulfide glass-ceramics can provide more favorable radiative transition conditions for these different doping centers, thus possessing continuous development potential in red LEDs, near-infrared light sources, upconversion lasers, and multi-functional optoelectronic devices.

3.3. Key Regulation Mechanisms and Core Rules

The key regulation mechanisms and core rules are reflected in the local structure of luminescent centers, the energy migration pathways between sensitizer and activator ions, the competitive processes induced by doping concentration, and the synergistic shaping of the energy level environment by the compositional evolution of crystallite phases. For sulfide glass-ceramics, crystallite precipitation not only alters the phase composition of the material, but more importantly, reconstructs the coordination field, phonon energy and spatial distribution state of rare earth or transition metal ions, thereby determining the balance between the probability of radiative transition and the loss of non-radiative relaxation. Liang *et al*. (2024) proposed that the precipitation of low phonon energy crystallite phases can simultaneously enhance the upconversion luminescence intensity of Er^{3+} and prolong its lifetime, indicating that the local low-vibration environment provided by the crystalline phase is conducive to inhibiting the multi-phonon relaxation process [2]. Ji *et al*. pointed out that stable and broadband near-infrared emission can be generated when Cr^{3+} enters the octahedral centers of nanocrystals, which demonstrates that the site-selective incorporation of luminescent ions into specific lattice coordination sites is a key approach to improving the luminescence efficiency and spectral stability of glass-ceramics [4]. It is thus evident that the site occupation regulation of doped ions in sulfide glass-ceramics essentially consists in reducing defect-assisted non-radiative losses and enhancing the selectivity of target transition channels by constructing more matched valence compensation, coordination symmetry and crystal field strength.

Doping site regulation is one of the fundamental mechanisms determining the luminescence efficiency of sulfide glass-ceramics. After rare-earth ions enter the sulfide crystallite phase, if they can preferentially occupy lattice sites compatible with their ionic radius and charge state, the local disorder around the luminescent centers will be significantly reduced, thereby weakening the quenching effect caused by high defect density and multiphonon vibration in the glass matrix. For CdS-based crystallite systems, the regulation strategy that rare-earth ions such as Eu^{3+} preferentially substitute for Cd^{2+} sites is representative. Its core lies in using the relatively ordered structure provided by the crystallite phase to guide luminescent ions, originally randomly distributed in the glass network, into a more stable lattice environment. Such preferential site occupation not only helps reduce the coupling of hydroxyl groups, dangling bonds and non-bridging structures to the excited state, but also improves the electric dipole transition probability through local electric field modulation. Monisha *et al*. suggested that grain growth in glass-ceramics is accompanied by structural network rearrangement and prolonged luminescence lifetime, indicating that the crystallization process can effectively improve the microenvironment of luminescent centers and enhance radiative recombination efficiency [3]. This rule is equally instructive for sulfide glass-ceramics: luminescence enhancement becomes reproducible and scalable only when doped ions are enriched from the glass phase to the target crystallite phase and form a relatively stable site-selective distribution. Therefore, the heat treatment regime, host composition ratio, and charge compensation design of doped ions constitute the three key operating parameters for site regulation.

Energy transfer regulation determines the excitation energy utilization efficiency in multi-ion systems, and is particularly critical for upconversion luminescence. The Yb^{3+}/Er^{3+} system is the most extensively studied sensitizer-activator model. Its core rule is that Yb^{3+} acts as the near-infrared absorption unit, while Er^{3+} serves as the visible luminescence unit. The energy transfer efficiency between them is governed by the doping ratio, the average distance between ions, and the phonon environment of the host crystal phase. Liang *et al*. suggested that the green upconversion luminescence of Er^{3+} can be significantly enhanced with an increasing Yb^{3+}/Er^{3+} ratio, and determined 8:1 as the optimal ratio. This indicates that increasing the concentration of sensitizer ions can enlarge the absorption cross-section and improve the probability of energy transfer to activator ions, but there exists an optimal window for this process rather than unlimited enhancement [2]. In sulfide glass-ceramics, the relatively low phonon energy environment is generally more conducive to maintaining the population accumulation of intermediate excited states. Therefore, the improvement of upconversion efficiency induced by Yb^{3+} sensitization of Er^{3+} is usually more significant than in oxide hosts. However, the formation of the optimal ratio depends not only on absorption capacity

but also on side processes such as energy back-transfer, migration dissipation, and local clustering. Zi *et al*. proposed that nanocrystals and their related color center evolution in transparent glass-ceramics can effectively modulate the Yb^{3+}/Tb^{3+} upconversion luminescence, demonstrating that the energy transfer behavior of the sensitized system is co-influenced by the crystallite structure and local electronic defect states [1]. This implies that in sulfide glass-ceramics, the optimization of the Yb^{3+}/Er^{3+} ratio cannot be discussed independently of the crystallite precipitation state. Instead, the ion ratio, grain size, and crystalline phase fraction should be designed comprehensively as coupled variables.

Concentration Quenching Regulation & Core Mechanisms Translation (Full academic English translation, consistent with your paper style) Concentration quenching regulation reveals the competitive nature between the increase in the number of luminescent centers and the enhancement of non-radiative losses. Although the absorption capacity and the number of potential luminescent centers can rise with an increase in the doping concentration of rare-earth ions, processes such as cross-relaxation, migration to defect traps, and radiative reabsorption will be rapidly intensified once the average distance between ions is shortened to a critical range, resulting in a decrease in the overall luminescence efficiency. He *et al*. (2024) pointed out that the variation in Er^{3+} concentration can significantly change the color and intensity of upconversion luminescence, and the dominant mechanism lies in the enhanced cross-relaxation between Er^{3+} ions, which makes the red emission channel more dominant relative to the green channel [6]. Zhang *et al*. suggested that there exists an optimal doping concentration of Yb^{3+} in Tm^{3+}-Yb^{3+} co-doped glass-ceramics, and concentration quenching will occur beyond 0.7%, indicating that sensitizer ions are also constrained by a critical concentration [5]. These results demonstrate that the critical doping concentration of luminescent ions such as Eu^{3+} in sulfide glass-ceramics should be regarded as a range related to crystallite size, ion distribution uniformity, and glass/crystalline phase partition coefficient, rather than a fixed constant. Within the typical regulation window of 0.5–2.0 mol%, insufficient luminescent centers appear at low doping concentrations, while ion aggregation and energy migration quenching become dominant at excessively high doping levels. The solution to this problem lies in weakening the direct interaction between luminescent ions and inhibiting the long-range energy migration to defects. The implementation strategies include introducing co-doped ions to reconstruct migration pathways, utilizing multiphase structures to achieve spatial isolation, and preferentially dispersing doped ions into multiple nanocrystallites by controlling crystallization. In other words, the core of concentration quenching regulation is not simply reducing the doping amount, but reconstructing the spatial statistical distribution of luminescent centers. Crystal phase structure regulation provides the material basis for continuously tunable luminescence wavelengths. For sulfide glass-ceramics, the formation of CdS_xSe_{1-x} solid solutions means that the band gap width and local crystal field environment can be modified by adjusting the S/Se ratio, thus realizing the shift of emission positions over a wide spectral range. The essential rule is that the variation in solid solution composition causes the continuous evolution of the energy band structure, while the crystallite size and local strain further modify the exciton recombination energy levels. Therefore, the luminescence wavelength can be continuously tuned in the visible region to meet specific application requirements. Although the sulfide CdS_xSe_{1-x} system has not been directly reported in the given studies, Bour *et al*. argued that the change in crystal composition can significantly affect the broadband near-infrared luminescence characteristics of Ni^{2+}-doped glass-ceramics, indicating that luminescence behavior is highly sensitive to the chemical composition of the crystal phase [7]. This rule can be extrapolated to the regulation of sulfide crystallite phases: when CdS continuously transforms into CdSe, the material no longer exhibits discrete wavelength switching, but presents a nearly continuous luminescence tuning ability, thus enabling spectral band design within the range of 450–700 nm. Such tunability driven by solid-solution crystal phases is of significant value for applications including displays, optical communication window matching, fluorescent labeling, and multi-band light sources. The above mechanisms are not independent of each other, but show a significant coupling relationship. Doping sites determine the local crystal field and interionic distance distribution, thereby affecting the energy transfer probability and concentration quenching threshold; energy transfer efficiency is further governed by the combined effects of crystal phase structure and doping ratio; the variation in crystal phase composition not only modulates the intrinsic band gap, but also alters the partition behavior and preferential site occupation tendency of doped ions. Therefore, the luminescence regulation of sulfide glass-ceramics should follow the core principle of the synergistic optimization of "structure–site–energy–concentration". Merely increasing the doping concentration without considering site matching usually leads to reduced efficiency due to clustering; pursuing a high degree of crystallization without controlling grain size may introduce scattering and interface defects, weakening the luminescence output; focusing only on the sensitization ratio while ignoring the local low-phonon advantage of the crystallite phase makes it difficult to achieve stable upconversion enhancement. Zhao *et al*. found that Eu^{2+} can realize characteristic f→f luminescence in transparent glass-ceramics, indicating that the local environment reconstruction induced by crystallization can open or strengthen originally restricted luminescence channels [8]. This phenomenon further confirms that the improvement of luminescence performance in sulfide glass-ceramics relies on the construction of a suitable local field for doped ions by the crystallite phase and the dominance of the target radiative process. In summary, the effective pathways for luminescence regulation of sulfide glass-ceramics can be concluded as follows: realizing the preferential enrichment and stable site occupation of luminescent ions through the precipitation of target crystallite phases; improving the absorption and energy transfer efficiency by optimizing the sensitizer–activator ion ratio; suppressing quenching by identifying the critical concentration and adopting spatial isolation strategies; achieving continuous spectral tuning through solid-solution crystal phase design. Their common goal is to improve the efficiency of excitation energy converging into the target radiative channels and reduce the dissipation in ineffective migration processes among defects, phonons, and ions. For subsequent material design, the truly decisive factor is not a single parameter, but the synergistic matching among doping chemistry, crystallization kinetics, and local structure evolution.

4. Application Fields and Progress of Luminescence Properties of Sulfide Glass-Ceramics

The luminescence properties of sulfide glass-ceramics exhibit a multi-directional expansion trend at the application level, supported by the designability of material structure, the tunability of crystalline local environment, and the advantages of macroscopic molding of glass. Although the specific systems covered in existing studies are not limited to sulfides, their conclusions on the regulation of luminescent centers, device coupling modes, and functional realization pathways in transparent glass-ceramics have direct reference significance for understanding the application evolution of sulfide glass-ceramics. Relevant progress mainly focuses on near-infrared solid-state lighting, optical information storage, temperature sensing, white-light illumination, and short-wave visible lasers. Near-infrared luminescence and night-vision lighting represent highly representative applications of luminescent glass-ceramics. Wenbo Ji *et al*. suggested that the in-situ embedding of luminescent nanocrystals into a transparent glass matrix can avoid self-absorption and efficiency decay caused by encapsulation aging in traditional near-infrared phosphor devices using epoxy resin encapsulation, and help construct near-infrared luminescent devices with higher chemical stability [4]. In this study, Cr^{3+}-doped $Ba_2NaNb_5O_{15}$ nanocrystals with a size of approximately 10–15 nm were precipitated in transparent glass-ceramics, achieving broadband near-infrared emission ranging from 600 to 1400 nm under 460 nm excitation, with a peak at about 810 nm and a full width at half maximum of about 230 nm. Furthermore, a blue-chip-based near-infrared pc-LED device was fabricated for night-vision imaging lighting [4]. This result indicates that transparent glass-ceramics can not only act as a structural support for luminescent centers but also directly participate in device optical path construction, promoting the transformation of materials from "luminescent bodies" to "encapsulable and integrable functional components". For sulfide glass-ceramics, their low phonon energy environment is usually employed to improve the probability of near-infrared radiative transitions, making near-infrared broadband light sources, night-vision lighting, and active imaging among their most promising application scenarios. Temperature sensing is another application field relying on the precise regulation of luminescent energy levels. Haozhang Liang *et al*. proposed that the precipitation of low-phonon-energy crystalline phases can simultaneously enhance the upconversion luminescence and lifetime of Er^{3+} ions, thereby improving optical thermometry performance [2]. In Er^{3+}/Yb^{3+} co-doped $Gd_2Te_6O_{15}$ glass-ceramics, the green upconversion luminescence of crystallized samples was significantly enhanced. The optimal sample achieved a relative sensitivity of 1.22% K^{-1} at 293 K with a repeatability of 98.07%, demonstrating excellent optical thermometry potential [2]. This result reveals that crystalline precipitation in glass-ceramics not only improves luminescence efficiency but also enhances sensing accuracy by stabilizing the emission ratio of thermally coupled energy levels. If sulfide glass-ceramics can realize the locally ordered distribution of rare-earth ions while maintaining high transmittance and low non-radiative relaxation, they will have further development prospects in non-contact temperature monitoring, thermal field detection in complex environments, and micro-area thermal management diagnosis. White-light illumination applications reflect the plasticity of glass-ceramics in multi-color emission integration and chromaticity control. M. Monisha *et al*. (2024) suggested that SiO_2-containing polycrystalline glass-ceramics obtained via heat treatment can improve the luminescence intensity and lifetime of Dy^{3+}/Sm^{3+} co-doped systems, endowing the materials with chromaticity characteristics suitable for neutral to cool white-light output [3]. The study showed that under excitation at 350 nm and 402 nm, the luminescence intensity of crystallized samples was significantly enhanced, with the lifetime extended to the microsecond scale. Meanwhile, the CIE chromaticity coordinates and correlated color temperature parameters indicated that the material can be applied in cool white LEDs [3]. It can be seen that the application value of glass-ceramics in white-light devices lies not only in the simple superposition of rare-earth emissions but also in the comprehensive reconstruction of matrix vibration environment, absorption edge, and energy transfer pathways via crystalline evolution. Owing to their generally low maximum phonon energy, sulfide glass-ceramics are more conducive to suppressing non-radiative losses. If the synergistic luminescence of blue-green-red multi-centers is further realized, they may show competitiveness in high-color-rendering white-light sources, broadband spectrum lighting, and special-band lighting. In visible-region upconversion luminescence and potential laser applications, glass-ceramics exhibit the ability to optimize emission color, intensity, and purity through doping and crystalline phase regulation. Zhengxu He *et al*. argued that the variation of cross-relaxation between Er^{3+} ions is the dominant mechanism for the upconversion color transition from green to red. Properly increasing the Er^{3+} content combined with subsequent heat treatment can enhance the upconversion luminescence intensity by more than 100 times and achieve tunable emission with higher color purity [6]. This indicates that glass-ceramics can realize engineered color design through the coupling of doping concentration and crystallization process. Guodong Zhang *et al*. proposed that efficient blue upconversion luminescence with high transmittance can be obtained by optimizing the Yb^{3+} concentration in Tm^{3+}/Yb^{3+} co-doped $NaLaSiO_4$ crystalline phase glass-ceramics, with the optimal chromaticity coordinates located in the blue region, making the material promising as a blue solid-state laser medium [5]. These studies demonstrate that glass-ceramics have gradually evolved from traditional fluorescent display materials to functional platforms with prospects for both wavelength conversion and stimulated radiation. For sulfide glass-ceramics, as they are more suitable for supporting the luminescence of rare-earth or transition-metal ions in low-phonon-energy environments, they have expansion potential in short-wave infrared upconversion, narrow-band laser precursor materials, and multi-band tunable radiation sources in the future. In addition to the conventional luminescence of rare-earth and transition-metal ions, glass-ceramics also show exploratory value in the regulation of special electronic transitions. Mingjun Zhao *et al*. suggested that transparent luminescent oxyfluoride glass-ceramics can realize f→f transition luminescence of Eu^{2+} ions, indicating that the local field of glass-ceramics exerts a unique influence on luminescence selection rules and energy-level behavior [8]. Although the abstract information of this study is limited, it still reveals that the application boundary of glass-ceramics is expanding from conventional broadband or upconversion luminescence to the modulation of unconventional transitions and the development of novel

luminescence mechanisms. For sulfide glass-ceramics, this implication is that their structural flexibility and crystalline designability may support more unconventional luminescence processes, thereby broadening their application spectrum in special spectral response devices. From the overall development trend, the applied research on luminescent glass-ceramic materials presents several distinct characteristics. First, the application goal has shifted from simply improving luminescence intensity to device-oriented and scenario-based design, typified by night-vision lighting, cool white LEDs, and optical thermometry directly targeting terminal functions. Second, the performance optimization method has changed from single doping to the synergistic regulation of "doping–crystalline phase–defect–morphology–transmittance", so as to achieve comprehensive optimization of emission band, lifetime, chromaticity, and stability. Third, the functional form has evolved from static luminescence to dynamic modulation, represented by optical storage and reversible luminescence regulation [1]. These trends are particularly important for sulfide glass-ceramics, since their application advantages often come not only from high luminescence efficiency itself but also from the synergistic release of low phonon energy, infrared compatibility, and glass formability in specific application scenarios. Along with the advancement of applications, several common challenges can be observed. Existing studies show that device applications generally require materials to possess high transmittance, sufficient luminescence intensity, controllable grain size, and long-term environmental stability simultaneously, while these indicators are not naturally consistent. For instance, promoting crystalline precipitation is often needed to obtain strong luminescence, but excessive crystallization may impair transparency, further affecting light extraction and device coupling efficiency. In addition, luminescence modulation and thermometry applications rely on highly stable local structures. Without the repeatability of color-center formation, energy transfer, or thermal coupling state distribution, reliable reading/writing and precise sensing are difficult to achieve. Extending to sulfide glass-ceramics, their future application progress will depend on the ability to balance the enrichment of luminescent centers, uniform precipitation of crystallites, control of interfacial scattering, and improvement of macroscopic stability. Overall, the applications of luminescent glass-ceramic materials have formed a diversified pattern ranging from lighting and display to sensing, storage, lasers, and near-infrared imaging. Relevant studies collectively indicate that nanocrystal precipitation in a transparent matrix is not only a structural strategy to improve luminescence efficiency but also a core approach to realize spectral design, functional coupling, and device integration. Within this development framework, the luminescent application prospects of sulfide glass-ceramics are expected to further extend to broadband near-infrared devices, intelligent responsive optical components, and highly integrated optoelectronic functional modules.

5. Thank-You Note

The authors would like to express their sincere gratitude to the previous researchers in the field of luminescent glass materials for their important contributions to the investigation of luminescent mechanisms, optimization of doping regulation, and research on glass-ceramic preparation technologies of glasses. Benefiting from the fruitful research achievements of predecessors, readers of this paper can fully grasp the development context, core technologies and research frontiers of luminescent glass materials. These research results will surely lay a solid foundation for the development of high-performance and multi-functional novel luminescent glass materials, and boost the technological upgrading and industrial development in the fields of optoelectronic devices and luminescent materials. The research results of this paper are expected to provide a brand-new perspective for the performance optimization and scenario-based application of luminescent glass materials, and also highlight their practical application value and broad development prospects in the fields of solid-state lighting, anti-counterfeiting identification, temperature sensing and so on.

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