1

A Brief Introduction to Molecular Aggregates

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1.1 Introduction

1.1.1 Motivation

The opto-electronic materials act as the key components in many functional devices, as well as the detection and imaging systems, which can permeate every aspect of modern life. Their diverse applications and potential for innovation have garnered increasing attention, and the diverse properties in aggregated states enrich their applications, including data storage and encryption,[1,2] photovoltaic devices,[3–5] organic light-emitting diodes (OLEDs),[6–8] organic light-emitting transistors (OLETs) devices,[9,10] biological sensing and imaging,[11–13] etc. In these applications, the macroscopic performance of molecular aggregates is not just the simple overlay of single molecules. In many cases, the emergence of new behaviors is enabled by specific molecular packing arrangements. Likewise, the adjustability and uncertainty of molecular aggregation bring opportunities and challenges for designing high-performance opto-electronic materials. Until now, there is still no comprehensive theoretical system to illustrate the detailed structure-performance relationship. However, certain advantageous molecular arrangements and packing modes have been discovered in specific scenarios to guide the construction of efficient opto-electronic materials. As the investigation goes deeper and deeper, it is essential to study their properties from the molecular aggregation levels, in addition to the individual molecules. The purpose of this book is to explore the influence of single molecules and molecular aggregates on the properties of opto-electronic materials, providing feasible ideas for molecular design, thereby broadening the

application scenarios of opto-electronic materials, and improving the performance of corresponding detection or devices.

In the past several decades, the opto-electronic properties of organic materials have been primarily explored through the molecular design of functional groups and structural motifs. Molecules, as the smallest identifiable unit of the substance, have been the foundation for extensive research. Research based on molecular science has flourished, and a large number of organic opto-electronic materials have been developed.[14,15] However, some phenomena, such as the different luminescent behaviors observed in different polymorphic forms, remain difficult to be explained within a single-molecule framework. Molecular aggregates bridge the gap between single-molecule properties and the macroscopic opto-electronic performance of materials, providing more objective and perfect analysis methods. Accordingly, molecular aggregates are highlighted in these cases, thereby breaking the original single-molecule framework and gradually pushing the development of molecular aggregate science.[16–18]

Generally, organic molecules in extremely dilute solutions can be approximately considered as the single-molecule state by ignoring the influence of solvent molecules. As the concentrations increase along with the formation of molecular aggregates or precipitation, the overall properties in aggregated states change in most cases, compared to those in single-molecule states. The macroscopic properties in aggregated states often exhibit remarkable diversity and complexity.[19,20] Key factors such as intermolecular interactions and stacking modes are critical in determining these properties, necessitating a departure from a purely molecular perspective to a comprehensive framework for understanding structure-packing-performance relationships. This shift has profound implications for the development of opto-electronic materials, particularly in fields where luminescence properties play a central role (Figure 1.1).

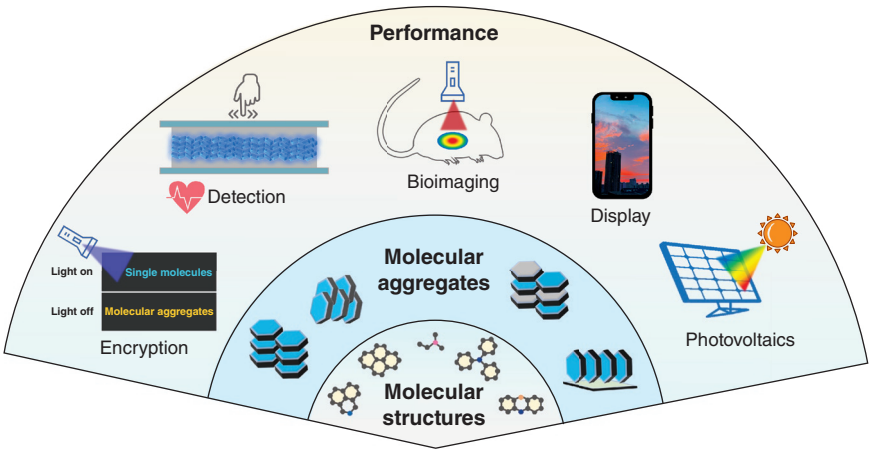

Figure 1.1 Schematic diagram of the relationship among molecular structures, molecular packing, and opto-electronic performance.

Luminescence properties can provide powerful and reliable information for exploring aggregated structures according to their changes from single molecules to aggregated states in some cases. Even for the same molecule, the tiny changes in aggregated structures can result in totally different luminescence properties, including emission wavelength, intensity, and lifetime. Moreover, specific phenomena such as mechanoluminescence (ML), mechanochromism, and room-temperature phosphorescence (RTP) often emerge from aggregated states, heavily influenced by molecular packing arrangements. Thus, in-depth exploration of the relationship between luminescence properties and aggregated structures represents a critical pathway to achieving excellent and diverse opto-electronic properties.[21–25]

The study of molecular aggregation and the related structure-performance relationship promotes the optimization of the photophysical properties and effectively improves the controllability of their opto-electronic property in the corresponding devices. For instance, in photovoltaic devices, the π–π coupling between two adjacent molecules is considered to be an efficient and important channel for charge transfer.[26–28] The research on the spatial structure of aggregated states can provide insights into the design of next-generation high-performance photovoltaic materials. For dye-sensitized solar cells, dye aggregation, which relates to the concentration of dye bath, ionic strength, temperature, solvents, and so on, usually leads to ineffective light-harvesting and charge recombination.[29] Accordingly, the efficiency of electron injections decreases, resulting in poor photovoltaic performance in most cases. Thus, the suppression of dye aggregation on the semiconductor surface is essential to improve the device performance. In perovskite solar cells, hole-transporting materials (HTMs) extract photogenerated holes and transport charges, thereby affecting the overall photovoltaic conversion efficiencies. The corresponding hole mobility and conductivity can be improved based on the optimization of morphology and crystallinity of hole-transporting layers (HTLs).[30] Although many effective strategies have been proposed, the relationship between molecular structure, aggregation behavior, and photovoltaic performance remains insufficiently understood. This limitation may be related to the complex carrier transfer processes and diverse molecular stacking in HTLs. Thus, precise molecular design guided by a deep understanding of aggregated structures is of great significance for improving device performance.

Similarly, the bio-applications of organic luminescence materials also depend on their aggregation behaviors under physiological environments, encompassing fields including biosensing, bioimaging, and therapy. For instance, aggregation-induced emission (AIE) luminogens and RTP nanoparticles demonstrate remarkable potential in biomedical imaging and therapeutic applications in aggregated states.[31,32] RTP materials can keep emissive states after turning off the light source, which shields the autofluorescence of tissues *in vivo*, thus achieving afterglow imaging of targeted tissues with high signal-to-noise ratios. In some cases, the reactive oxygen species (ROS) can be generated under external stimuli, including light and ultrasound, which can effectively kill tumor cells through photodynamic therapy or sonodynamic therapy.[33,34]

However, a lack of comprehensive analysis of molecular packing in opto-electronic materials still results in an increased gap between molecular design and practical applications. Although some dominant molecular packing modes have been studied in past research, several factors still hinder a systematic and deep understanding. On the one hand, the methods for characterizing and testing molecular aggregates are still limited, making it hard to monitor molecular aggregation behavior in practical applications. So far, the analysis of aggregated structures is heavily dependent on single crystals. However, in specific applications, molecular aggregation often takes more complex forms, including films, nanoparticles, powders, etc. At present, the intermolecular interactions and molecular stacking can be predicted through theoretical calculation, but discrepancies frequently arise between simulations and experimental results. On the other hand, the relationship among molecular structures, aggregation behaviors, and opto-electronic properties is still not clear enough to guide the practical application. Although some useful strategies have been proposed and applied in relatively simple systems, extending these approaches to complex molecular structures and multi-component systems across diverse opto-electronic fields remains a significant challenge. Thus, research on the relationship among molecular structures, aggregated modes, and opto-electronic properties is essential, and a mesoscale perspective bridging the gap between molecular design and material application is urgently needed.

1.1.2 A Brief History

The research and exploration of aggregated modes in organic opto-electronic materials has been one of the most advanced fields. The journey from molecular monomers to aggregated states has witnessed groundbreaking milestones since the first introduction of these organic opto-electronic materials, involving photoluminescence (PL), ML, OLEDs, OLETs, nonlinear optics (NLO), organic photovoltaics (OPV), perovskite solar cells, photocatalytic hydrogen evolution, and so on.

Luminescent materials, in particular, exhibit highly sensitive optical signals that enable rapid and pronounced responses from monomer to aggregate states, significantly influencing the development of modern opto-electronic devices and imaging technologies. It can be divided into fluorescence and phosphorescence based on the pathways of irradiative decay from singlet or triplet excited states. Since the nineteenth century, Sir John Herschel first observed that the colorless solution of quinine displayed a beautiful celestial blue color under light,[35] various organic fluorogens have been developed.[36] In 2001, Tang et al. proposed the concept of AIE from an unusual phenomenon that the non-emissive solution of 1-methyl-1,2,3,4,5-pentaphenylsilole became fluorescent upon aggregation on a thin-layer chromatography plate.[37] The emergence of the intriguing AIE effect established a new era of aggregated luminescent molecules, and AIE materials have since been widely applied in bioimaging and opto-electronics,[38,39] supported by a growing understanding of their underlying mechanisms.[40] Currently, a universal database for aggregate science has been established to facilitate a data-driven research paradigm within the field.[41]

Compared to fluorescence, phosphorescence refers to the emission of photons from the triplet excited states. Due to the inherent instability of the triplet states, phosphorescence is highly sensitive to the external environment and strongly dependent on its own aggregated modes. In 1939, the first observation of organic room-temperature phosphorescence in the solid state was reported by Clapp using the crystalline tetraphenylmethane and tetraphenylsilane derivatives.[42] The continuous discovery of the phosphorescence phenomenon advanced the corresponding theories. In the 1970s, Parker et al. adsorbed sodium 4-biphenylcarboxylate and sodium 1-naphthoate on paper, sucrose, starch, glass fiber paper, and silanized paper, and proposed the prototype of the concept that the restriction of molecular motions was favorable to RTP performance.[43] More recently, the emergence of crystallization-induced phosphorescence (CIP), reported by Sun and Tang et al. in 2010, demonstrated that ordered and compact packing with multiple intermolecular interactions could inhibit the possible quenching of the triplet state.[44] Since then, organic RTP materials with high efficiency in aggregated states have been gradually developed, and the relationship between molecular packing and RTP properties has been underscored.[45] In 2018, Pu and Li et al. studied the effects of the molecular packing on the organic RTP of the 10-phenyl-10H-phenothiazine-5,5-dioxide-based derivatives,[46] and elucidated the important role of π–π coupling in the stabilization of triplet excitons. Until now, the effect of molecular packing on RTP has been systematically investigated, highlighting the close relationship between aggregation modes and phosphorescent properties.[47]

ML materials, characterized by light emission induced by mechanical stimuli such as rubbing, scratching, pressing, or shaking, also show a strong dependence on their aggregated state. The first discovery of ML traces back to 1605, when Bacon observed light emission by scraping hard sugar crystals.[48] Over the next several centuries, various ML materials were reported, such as inorganic crystals, minerals, and organic crystals, including phenanthrene, 9-anthrylethanol, coumarin, acenaphthene, carbazole derivatives, *N*-phenyl imides, and so on.[49] With the development of AIEgens, a series of ML-AIE active molecules were reported in the 2010s,[50] which facilitated the understanding of ML mechanism and the exploration of the relationship between aggregated structures and ML. However, these ML materials usually exhibited only fluorescence as short-lived luminescence. In 2017, Li et al. reported a mechanoluminescent material based on phenothiazine derivatives with dual emission of fluorescence and phosphorescence.[51] More recently, Liu et al. reported ML materials by isostructural doping strategy, making the ML emission with an extended duration of up to 0.4 seconds.[52]

The advancement in aggregate science also facilitates the development of electroluminescence (EL) materials, which emit light in response to the passage of an electric current or a strong electric field. OLED devices based on EL materials have become a cornerstone in modern opto-electronics. The origins of OLED technology trace back to the early 1960s, with the discovery of delayed fluorescence in eosin and EL in anthracene crystals under high voltage.[53] While traditional fluorescent materials suffered from aggregation-caused quenching effect with limited efficiencies in OLEDs, in 2001, Tang et al. found the AIE effect in compound

1-methyl-1,2,3,4,5-pentaphenylsilole.[37] Since then, AIEgens with twisted molecular structures like triphenylamine, tetraphenylethylene, 10H-spiro[acridine-9,9'-fluoren] have been widely used in the non-doped OLEDs.[54,55] Meanwhile, researchers continuously investigated new OLED materials based on phosphorescence and thermally activated delayed fluorescence (TADF).[56] The aggregation-induced delayed fluorescence (AIDF) principle proposed by Chi and Tang et al., with the combination of AIE and TADF,[57,58] has further enhanced OLED performance. AIDF-based OLEDs have achieved unprecedented external quantum efficiencies (EQEs) by maximizing exciton utilization through aggregation.

Extending beyond luminescent materials to other critical applications, the research of aggregate science could be traced back to some other opto-electronic materials, without the consideration focusing on aggregate science at that time.[59–62] For example, as a significant branch of modern optics, nonlinear optics also exhibits a strong dependence on molecular packing modes. Indeed, early breakthroughs in this field originated from crystalline materials. In 1961, Franken et al.[63] made a landmark discovery and observed the occurrence of second harmonic generation (SHG) in a ruby crystal. In 1964, Rentzepis et al.[64] detected SHG in organic crystals such as benzopyrene, marking the early steps toward organic nonlinear optical materials. By 1970, Davydov et al.[65] reported that organic materials with electronic push-pull structures exhibited second-order nonlinear optical responses, utilizing intramolecular charge transfer to trigger nonlinear optical activity. In 1999, Dalton et al.[66] introduced the "site isolation principle," where chemical modifications were used to change chromophore spatial conformation toward a spherical shape, thus weakening strong dipole–dipole interactions and improving macroscopic NLO effects. Guided by this principle, dendrimers have emerged as promising NLO materials since they can protect chromophores and reduce dipole–dipole interactions among them through dendritic isolation groups.[67,68] The transition from single molecules to macroscopic systems, such as thin films, reveals several challenges and opportunities related to molecular packing and aggregation states. A clear discrepancy exists between the microscopic NLO effects of chromophores and the macroscopic second-order NLO effects of thin films. At the microscopic level, nonlinear polarization arises from electron transitions induced by photon absorption. However, in macroscopic systems, dipole–dipole interactions and the orientation of molecules significantly influence the overall NLO response. Significant progress has been made in fine-tuning molecular aggregation to optimize NLO materials by the incorporation of site isolation principle. From 2006, Li et al.[69,70] introduced various isolation groups to facilitate chromophore alignment during polarization, and the concept of "suitable isolation group" was further proposed to achieve spatial arrangements that facilitated orderly molecular alignment and enhanced NLO performance.[71–73]

The regulation of molecular aggregation to the improved performance of photovoltaic devices is equally groundbreaking.[74–77] For perovskite solar cells, the aggregation characteristics of organic semiconductors have long been recognized as a critical factor in enhancing the carrier transport capabilities of organic semiconductor-based hole transport materials (HTMs).[78,79] Photocatalytic hydrogen

evolution has also benefited from the strategic manipulation of aggregation behavior. Upon rational aggregation of chromophores, intermolecular interactions promote the emergence of new coupled states that mitigate the Coulombic attraction between electrons and holes, thus enhancing charge separation and the subsequent hydrogen evolution.[80,81] The size of aggregates is a critical determinant in photocatalytic performance. It dictates the specific surface area of active sites and electron transport pathways. Meanwhile, aggregates with higher crystallinity and fewer recombination centers provide an ideal environment for photogenerated carriers to diffuse more effectively from the bulk to the surface, where redox reactions take place.[82]

In conclusion, the above discussions demonstrate how aggregate science has reshaped the design and applications of opto-electronic materials. By understanding and controlling molecular packing and aggregation states, researchers have unlocked unprecedented levels of performance in opto-electronic materials. These advancements not only highlight the versatility of aggregation phenomena but also provide a robust foundation for future innovations in molecular design and opto-electronic technologies.

1.1.3 Basic Knowledge of Organic Compounds in Aggregated States

The previous sections introduced the motivation and development of molecular aggregated states. The following content explains some common elements of molecular aggregate science, focusing on the driving forces behind molecular aggregation and their profound influence on the macroscopic properties of functional materials. These aspects are critical for understanding and regulating the aggregated states.

1.1.3.1 The Driving Force of Molecular Aggregates

The aggregated behaviors of organic molecules are derived from various intermolecular noncovalent interactions, involving hydrogen bonds, π–π interactions, electrostatic interactions, hydrophobic associations, and so on. In contrast to single covalent bonds with energies higher than 200 kJ mol^{-1}, noncovalent interactions are relatively weaker but highly significant in driving molecular aggregation. For example, hydrogen bonds exhibit energies of 25–40 kJ mol^{-1}, while the energies of other noncovalent interactions are lower than 10 kJ mol^{-1}.[83–86] These interactions act as the molecular linker, combining individual molecules to form macroscopic materials with diverse functional properties (Figure 1.2a), where molecular aggregates influence charge transport, luminescence, energy conversion efficiency, etc.[40,45,87] Broadly, these noncovalent interactions can be categorized as follows.

1.1.3.1.1 Hydrogen Bond

Hydrogen bond, one of the most extensively studied noncovalent interactions, involves a hydrogen atom bonded to a highly electronegative atom that interacts with a nearby electronegative atom with lone electron pairs, resulting in a special noncovalent interaction in the form of X—H···Y (Figure 1.2b). X and Y represent non-metallic atoms with high electronegativity and small atomic radii, such as

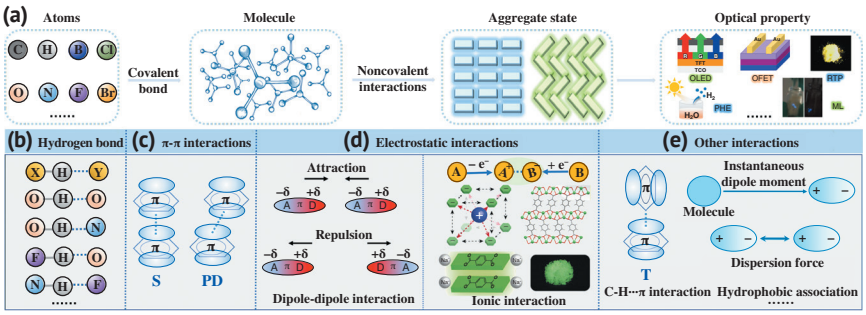

Figure 1.2 (a) Schematic diagram of molecular aggregated state formation. (b) Hydrogen bonding and the representation. (c) π–π interactions and the representation. (d) Electrostatic interactions and the representation including dipole–dipole interactions and ionic interactions. (e) Other interactions and the corresponding representation.

F, O, N, etc.[88,89] Hydrogen bonds can form between identical atoms, such as O—H···O in water, or between different atoms, like N—H···O in aqueous ammonia. Moreover, a hydrogen bond is characterized by specificity, saturability, and directionality, with optimal strength achieved when X—H and Y are aligned linearly, although it can also be bent or bifurcated. No matter the binding energy of a single hydrogen bond is relatively lower than that of covalent bonds, the cumulative effect of hydrogen bonding is significant.[90–92] In molecular aggregates, multiple hydrogen bonds often work synergistically to stabilize the aggregated structures.[93] This is particularly relevant in organic opto-electronic materials like hydrogen-bonded organic frameworks as chemical sensing materials and molecular devices, where strong hydrogen-bonding networks contribute to the thermal and mechanical stability of materials, improving their device performance.[94–96]

1.1.3.1.2 π–π Interaction

The π–π interaction is another pivotal one among aromatic systems, which is driven by dispersion forces between π-electron clouds. It is crucial for efficient charge transport and exciton diffusion in devices such as OLEDs and OPVs.[97–100] Depending on the molecular geometry, π–π interactions can occur in different stacking modes, such as S-shaped (sandwich) and PD-shaped (parallel-displaced) stacking (Figure 1.2c).[101–103] Understanding the nature of π–π interactions is essential for regulating the opto-electronic properties of organic materials, such as light absorption, charge transport, emission properties, etc. For instance, in pentacene derivatives, S-shaped stacking enables high charge-carrier mobility by creating continuous π-conduction pathways.[104,105] PD-shaped stacking, often observed in conjugated polymers, enhances the optical and electronic properties of these materials.[106]

1.1.3.1.3 Electrostatic Interactions

Electrostatic interactions add another dimension to the stabilization and functionality of molecular aggregates. These forces arise from the attraction between opposite dipoles or charged ions, including dipole–dipole and ionic interactions.[107–109] (Figure 1.2d). Ionic interactions occur between cations and anions, playing an

important role in improving structural rigidity and tuning the solubility and processing conditions of organic materials. Polar molecules exhibit uneven charge distributions with permanent dipole moments, resulting in the generation of dipole–dipole interactions between them. Therefore, the interactions are created between partial positive and negative charges that attract one another. In opto-electronic materials, electrostatic interactions play a crucial role in improving the overall performance of opto-electronic devices by fine-tuning their arrangements. For example, in organic materials used in OPVs, electrostatic forces can facilitate the formation of phase-separated domains that optimize exciton dissociation and charge collection efficiency.[110,111]

1.1.3.1.4 Other Interactions

In addition to π–π and electrostatic interactions, several other types of interactions contribute to the aggregation modes of molecular systems in opto-electronic materials. C—H···π interactions, for example, are essential in π-stacking systems, where a hydrogen atom with slight electron deficiency on one aromatic ring interacts with the π-electron cloud of another aromatic ring, beneficial to the formation of T-shaped stacking (Figure 1.2e).[112] These interactions can also facilitate the formation of tightly packed and stabilize the aggregated structures. For example, the herringbone packing mode in ML materials, driven by edge-to-face molecular arrangements with multiple C—H···π interactions, prevents non-radiative relaxation by minimizing molecular slippage under mechanical force, thus facilitating the generation of ML.[113,114] Furthermore, hydrophobic associations, though less intuitive, are vital in the self-assembly of nonpolar molecules in polar environments like water. These interactions arise because polar water molecules preferentially interact with each other, forcing nonpolar molecules to aggregate into hydrophobic domains. This phenomenon contributes to the formation of micelles and vesicles and plays a significant role in stabilizing organic molecules in aqueous dispersions.[115–118]

While permanent dipole moments of polar molecules result in the generation of dipole–dipole interactions, instantaneous dipole moments can lead to the generation of dispersion forces due to the fluctuating distribution of electrons within a molecule. They exist between all molecules and result in weak attractions when molecules come sufficiently close to each other (Figure 1.2e).[119,120] Quantum mechanical calculations reveal that the strength of dispersion forces is strongly related to the polarizability of molecules, and the more deformable or polarizable molecules exhibit stronger dispersion interactions.[121] In opto-electronic materials, dispersion forces play a pivotal role in governing the stability of molecular assemblies. These interactions help stabilize the molecular packing within crystals and influence the morphology of aggregated structures, which in turn affect the macroscopic properties. For instance, halogen bonds can significantly limit the non-radiative transitions of molecules and enhance SOC, contributing to the RTP properties of the material.[122,123]

The aforementioned interactions often act synergistically to determine the overall properties of molecular aggregates. For example, in organic thin films used in light-emitting materials, hydrogen bonds can stabilize the molecular framework, while

π–π stacking governs the packing density and electronic coupling between molecules. Electrostatic interactions further refine molecular orientation, enhancing the optical properties. In the design of materials for OFETs, precise tuning of π-π stacking and hydrogen bonding ensures high charge mobility and stability. Similarly, in organic bioimaging systems, hydrophobic interactions can help maintain the integrity of dye molecules in aqueous media, while hydrogen bonds, π-stacking, and electrostatic interactions can stabilize excitons and suppress non-radiative transitions, thereby enhancing the bioimaging performance.

1.1.3.2 The Adjustment of Molecular Interactions in Molecular Aggregates

Actually, the interplay of the driving forces of molecular aggregates, such as hydrogen bonding, π-stacking, electrostatic interactions, and hydrophobic associations, not only governs the formation and stability of molecular assemblies but also tailors their optical and electronic properties. The macroscopic functions of opto-electronic materials in aggregated states are usually different from those of single molecules, such as in dilute solutions, which are related to molecular packing in most cases.[20,124–126] In opto-electronic applications, the molecular packing of organic compounds is a critical factor influencing their properties, such as luminescence, charge mobility, and energy efficiency. Thus, controlling molecular packing is an essential issue for achieving the optimized optical and electronic properties. A comprehensive understanding of these interactions and the efficient strategies to regulate them will provide a powerful toolkit for designing innovative materials.

Though great challenges exist because of the unclear structures and complicated intermolecular interactions, advancements in molecular design have enabled the adjustment of molecular interactions and packing through internal and external forces (Figure 1.3). The internal forces include electronic and steric effects, the self-assembly effect, etc. The electronic properties of molecules can be manipulated to influence their packing behavior. Adjustments to the electron density of π-conjugated backbones or the electronic nature of substituents can regulate the degree of electronic coupling in aggregates, enabling diverse packing arrangements. For example, fine-tuning π-electron coupling through the regulation of substituents has been proven to be an effective strategy to regulate the RTP emission.[46] Similarly, substituents with electron-donating or electron-withdrawing characteristics can modulate the alignment of molecules and the electron affinities of materials, tailoring the electron mobility and charge transport properties of opto-electronic materials.[127]

The steric effect provides another strategy to adjust molecular packing by influencing spatial arrangement. When substituent groups with various sizes or shapes are integrated into molecular backbones, molecules will be dictated to adopt a compact or loose arrangement, leading to different optical and electronic properties.[128,22] Accordingly, researchers often maintain the main structures of molecules and change the substitutes, such as introducing specific functional groups or alkyl chains, to realize the subtle adjustments in molecular packing modes and associated performance.[129,130] The introduction of bulky substituents has been demonstrated to effectively disrupt π–π stacking, and the odd-even effect of alkanes has been widely utilized for the regulation of the molecular packing and intermolecular interactions.[131]

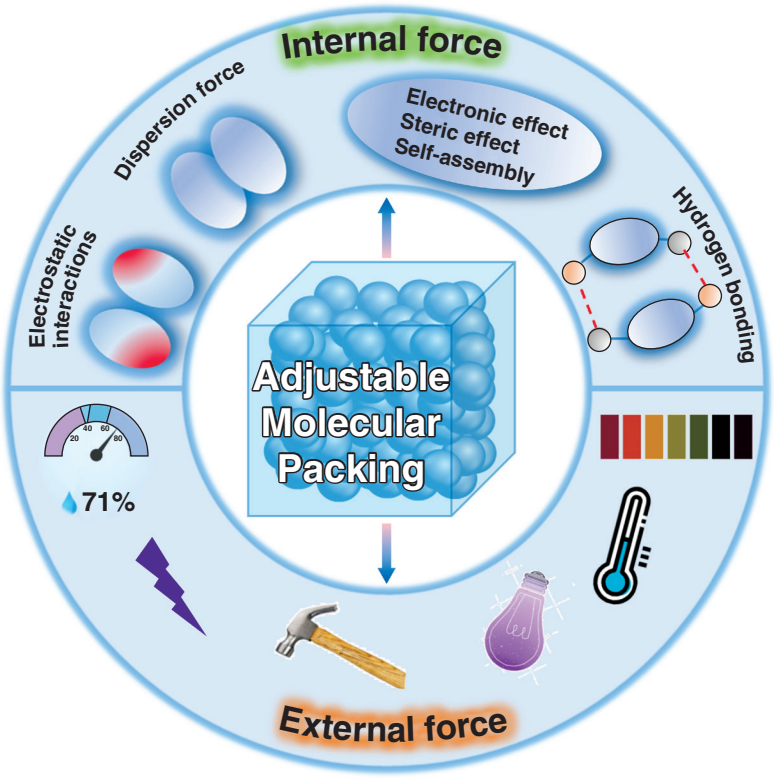

Figure 1.3 Schematic diagram of adjusting molecular packing through internal and external forces.

Self-assembly is an intrinsic mechanism that enables molecules to organize into well-defined aggregates without external guidance. This process is largely governed by noncovalent interactions, which can be modulated to achieve desired packing modes. It arises from balances of intermolecular forces, including hydrogen bonding, van der Waals forces, π–π interactions, dipole–dipole interactions, and hydrophobic effects. These interactions are finely tuned to achieve thermodynamically stable or metastable aggregates with desired functionalities.[132] The advantage of self-assembly lies in its ability to amplify intermolecular interactions and guide the spatial arrangement of molecules without the need for extensive manipulation. For instance, assembly systems with specific packing geometries can be realized by altering functional groups and the formation of hydrogen bonding.[133]

Moreover, external stimuli offer dynamic control over molecular aggregates, enabling reversible or irreversible transformations in their packing and properties. Upon the introduction of some external force, such as force, light, heat, electronic field, and others, dynamic optical properties can be achieved. Mechano-stimulation, including rubbing, stretching, or compressing, can induce changes in intermolecular interactions and alignments, disrupt crystalline packing, or alter molecular conformations. For example, the planarization of molecular conformations or the transition from

crystalline to amorphous states under mechanical stress can lead to mechanochromic effects or mechanoluminescence phenomena, yielding applications in stress sensors.[134,135] Mechanical stretching or compression can induce molecular alignment along the direction of the applied force. These alignments improve the corresponding properties, such as charge carrier mobility, birefringence, or optical transparency. Shearing forces, such as those applied during solution processing or coating, can orient molecular assemblies in specific directions. This is particularly effective for achieving uniaxial alignment in liquid crystals or polymer films.[136,137] Furthermore, the physical rubbing of a substrate using a specific tool (such as a soft cloth or a rubber roller), creates microscopic grooves on the surface. These grooves serve as topographical cues that guide the orientation of the molecules that are subsequently deposited onto the surface, causing them to align in the direction of the rubbing motion. This alignment can be transferred to the liquid crystal molecules deposited on top of the rubbed surface, which further align due to molecular interactions. It is widely used to control molecular orientation in liquid crystal.[138]

As for the photo-stimulation, some mechanisms are involved. Photo-stimulation has been found to utilize light to trigger molecular changes through processes such as isomerization, photodimerization, and photo-induced charge transfer. These processes can alter molecular geometry, dipole moment, or electronic distribution, which in turn modulate how molecules interact and arrange. Photoisomerization involves a reversible change in molecular conformations upon light irradiation.[19] For example, azobenzene switches between *trans* and *cis* forms under UV and visible light, respectively. This conformational change can alter molecular alignment and packing density, influencing properties such as charge transport and refractive index.[139] Certain molecules, such as anthracenes and coumarins, undergo light-induced dimerization. This process can create covalent bonds between molecules, driving them into specific packing arrangements or disrupting preexisting assemblies.[140,141] Photochromic molecules, such as spiropyrans, exhibit a reversible transformation between two states with different electronic and structural properties.[142] This enables light-triggered changes in molecular packing, orientation, or aggregation behavior. Besides, light could influence molecular packing by enhancing π-π interactions or suppressing quenching effects through oxygen consumption.[143]

Heat and electric fields play crucial roles in the modulation of molecular aggregation. Thermal energy provides the necessary activation for molecular systems to overcome energy barriers and reorganize their packing. This process is often governed by thermodynamic principles. The effects of heating processes include the increase of molecular mobility, phase transitions, and annealing of crystalline or semi-crystalline structures. Electric fields interact with molecular dipoles, charges, or polarizabilities, inducing reorientation and alignment. This mechanism leverages the ability of electric fields to exert torque or directional forces on polarizable molecules, promoting anisotropic packing and alignment.[144] Molecules with permanent or induced dipoles can align parallel to an applied electric field, creating ordered domains. This effect is widely used in liquid crystal technologies.[145,146] Heat and electric fields offer complementary and versatile tools for adjusting molecular packing and orientation, which are beneficial for the achievement of the desired orientation in materials such as

second-order non-linear optical and liquid crystal materials.[147,148] In these materials, thermal energy facilitates mobility and phase transitions, while electric fields provide directional alignment and polarization control.

Chemical stimuli could also change molecule packing, which affects molecular systems through a variety of mechanisms, including solvation, selective interactions, and chemical reactions. These interactions can alter the thermodynamic and kinetic pathways of molecular organization, enabling the precise tuning of packing density, phase behavior, and orientation. Selective solvents can promote specific interactions, guiding the self-assembly of molecules. These solvents can influence the solubility and molecular interactions of solutes, providing a thermodynamic driving force for specific rearrangement pathways.[149,150] The addition of specific ligands, ions, or templates can trigger supramolecular assembly driven by hydrogen bonding, coordination, or host-guest interactions.[151,152] Adjusting pH or ionic strength can influence charge distribution and electrostatic interactions in polyelectrolytes or ionic materials, enabling controlled orientation and packing.[153,154] By utilizing internal and external forces, these strategies continue to promote the development of next-generation devices, highlighting the critical intersection of molecular aggregate science in opto-electronics.

1.1.4 Overview of Topics Covered

Molecular aggregation plays a pivotal role in the functional properties and performance of opto-electronic materials. The proposed concept of "Molecular Uniting Set Identified Characteristic (MUSIC)" highlights the key role of molecular uniting in aggregated states. The exploration of molecular aggregates provides a profound understanding of their behaviors and potential in diverse applications, urging further research into their structure–property relationships. This section focuses on the key role of molecular aggregates in opto-electronic materials, including RTP, ML,[155–157] mechanoresponsive luminescence (MRL),[134,158,159] hole-transporting layer in perovskite solar cells, active layer in organic photovoltaics, OLED devices, and second-order non-linear optical materials. Their functionalities are closely related to the molecular aggregates,[24,45,16] including molecular packing and orientation, intermolecular interactions (hydrogen bond, π–π interactions, electrostatic interactions),[160] molecular configurations and conformations.[161] The diverse aggregation states of molecules such as crystals, co-crystals, thin films, nanoparticles, and fluids, exhibit distinct structural and functional characteristics, influencing their applicability in various advanced technologies.[162–164] The strategic design and control of these aggregated states are indispensable for achieving tailored opto-electronic properties (Figure 1.4).

Crystal or crystalline states represent highly ordered structures that exhibit reliable molecular packing and conformation for studying opto-electronic properties. Cocrystals consist of two or more components that form a unique crystalline structure. The precise packing modes and molecular conformations of both crystals and cocrystals can be characterized by single crystal/powder X-ray diffraction analysis, affording a convenient way to get insights into the possible relationship between

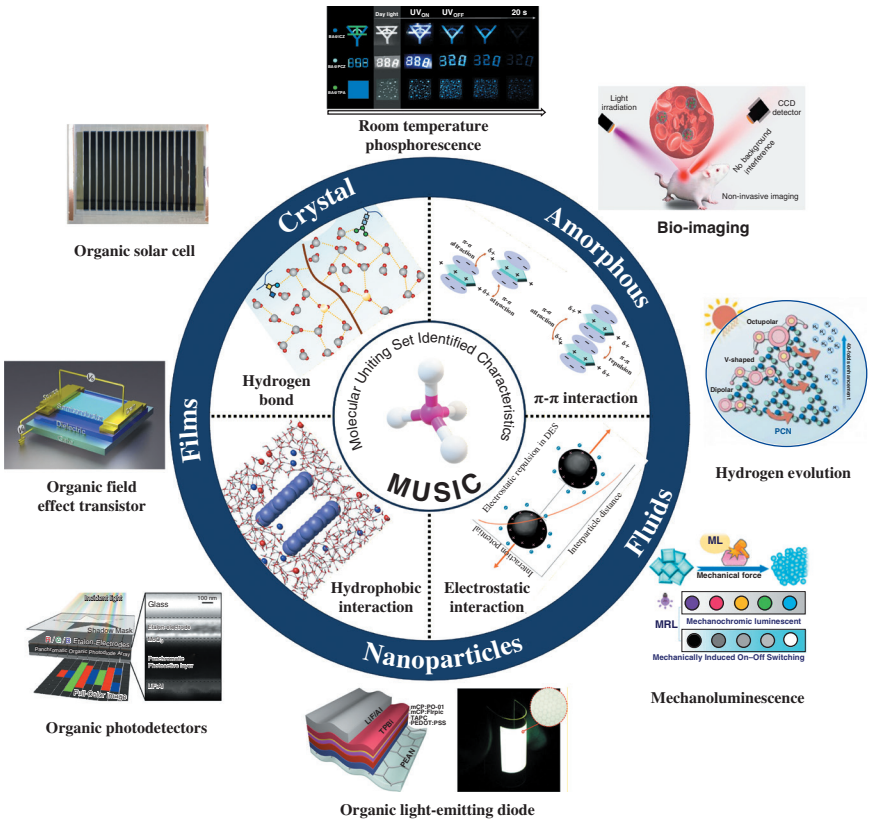

Figure 1.4 Schematic illustration of the possible aggregation states of molecules – crystals, films, nanoparticles, and fluids – to achieve applications in various fields. *Source*: Reproduced with permission from [165], Copyright 2023, American Chemical Society; [166], Copyright 2025, Royal Society of Chemistry; [167], Copyright 2007, American Chemical Society; [168], Copyright 2021, American Chemical Society; [169], Copyright 2020, American Chemical Society; [170], Copyright 2023, American Chemical Society; [171], Copyright 2024, American Chemical Society; [172], Copyright 2021, American Chemical Society; [173], Copyright 2022, Elsevier; [45], Copyright 2020, American Chemical Society; [174], Copyright 2014, American Chemical Society; [175], Copyright 2019, American Chemical Society.

molecular arrangement and opto-electronic properties.[158,176–180] For example, the herringbone packing mode with the ability to effectively suppress molecular slippage has been proven to be essential to ML crystals.[155,181] Controlled dimer formation in phenothiazine derivatives[182], anthracene derivatives and pyrene derivatives.[157,183] plays a key role in MRL materials. π–π coupling has been found to stabilize excited triplet states, enhancing RTP performance.[184–189] In other fields, such as the hole-transporting layer in perovskite solar cells, active layer in organic photovoltaics, and thermally activated delayed fluorescence (TADF) materials, molecular packing influences their charge transport and exciton behaviors, providing indirect evidence to explain the mechanism and guiding the material design.[176,190,191,30] However, it is not easy to get perfect crystals to fit the requirements of many opto-electronic

devices, and most crystal-based organic materials face challenges in processability, flexibility, and reproducibility, limiting their application in practical devices.[192]

Alternatively, thin films with amorphous or partially crystalline states, ranging from fractions of a nanometer (monolayer) to several micrometers in thickness, provide diverse functional properties in opto-electronic applications due to their ease of processing and adaptability to device integration. These films can be fabricated through spin coating, vapor deposition, and solution casting, enabling controlled aggregation and tunable functionality. For example, in dye-doped polymer films,[193] the aggregation behaviors can be controlled by changing the concentrations of dyes and the temperatures in the solution casting process, and the emission from different triplet states can be achieved, enabling adjustable afterglow colors. In stimulus-responsive copolymer films,[194–196] rigid cross-linked polymer network and hydrogen bonding among polymer chains effectively suppress the motion of polymer chains, and prevent the quenching of triplet excitons from oxygen and moisture, enabling ultralong RTP emission.

In organic semiconductor layer of OFETs, molecular aggregation is equally vital. The introduction of additives promotes electrostatic interactions that facilitate dynamic nucleation and crystal growth, resulting in long-range ordered crystalline domains. These ordered domains enhance charge transport, leading to high charge-carrier mobility, which is critical for device performance.[169,197,198] For active layers in polymer light-emitting diodes,[199,200] hole-transporting layers in perovskite solar cells,[30,201,202] and active layers in hybrid solar cells,[191,203,204] aggregation and morphology significantly affect exciton dissociation and carrier transport properties, and change the performance of devices accordingly,[205–209] which can be achieved through the optimization in film preparation processes. Thin films offer a practical alternative to crystalline materials, balancing structural randomness with functional performance while addressing the challenges of scalability and flexibility.

A nanoparticle or ultrafine particle usually exhibits the dimension ranging from 1 to 100 nanometers (nm) in diameter. The term is sometimes used for larger particles up to 500 nm in diameter, or fibers and tubes that are smaller than 100 nm in only two directions. They exhibit unique opto-electronic properties dictated by their size, morphology, and aggregated states. These properties can be finely tuned through controlled fabrication processes, including the modulation of nucleation, crystal growth, intermolecular interactions, and the ratio of orthogonal solvent or adding different surfactants. In the field of photocatalytic hydrogen production, by changing the solvent ratios in the fabrication process of nanoparticles, the morphology can be optimized effectively due to the fine tuning of molecular packing in aggregated states, and better photocatalytic performance can be achieved.[210,211] For healthcare, nanoparticles are one of the most popular aggregate states to be investigated in bioimaging, labeling, diagnosis, and treatment. Traditional fluorescent dyes, such as fluorescein, rhodamine B, or cyanine dye,[212] can help to solve problems including labeling and observation as nanoparticles. Until now, to satisfy the requirement of high signal-to-noise ratio, low toxicity, single targeting, long afterglow nanoparticles are designed and fabricated through a top-down preparation method. The selected compounds include carbazole,[213] phenothiazine,[214,51]

acridine,[215] xanthene,[216] triphenylamine,[217] phenylboronic derivatives,[218] and benzothiazole[219] derivatives. Requirements for fluorescent/phosphorescent materials in the applications in biomedical field keep increasing. For example, in the COVID-19 test, gold nanoparticles,[220,221] fluorescent nanoparticles (time-resolved fluorescence, fluorescent microspheres, and quantum dots),[222–224] magnetic nanoparticles, liposomes, rare earth nanomaterials, etc. have been developed to detect COVID-19 antigen due to the urgent need for effective detection.[225]

Furthermore, some nanoparticles demonstrate properties of stimulus-responsive due to the presence of specific groups. For instance, pH changes can promote the protonation or deprotonation, or the formation or breaking process of chemical bonds, while light irradiation can lead to isomerization or formation of chemical bonds due to the presence of some light-sensitive structures. Thus, environmental changes have a direct impact on intermolecular interactions in these nanoparticles, inducing morphological and functional transformations and enabling applications in diagnostics and responsive devices.

Fluids, including liquids and other nearly incompressible states of matter, represent a unique aggregate state with distinct physical and molecular characteristics. While fluids conform to the shape of their containers, they maintain a nearly constant volume regardless of external pressure, exhibiting fluidity, compressibility, and viscosity. These properties arise from the molecular dynamics in the liquid state, where molecules exhibit long-range disorder yet maintain short-range order. This semicrystalline behavior manifests as the local molecular organization that is transient and dynamic, distinguishing liquids from other aggregated states. Each molecule in liquid is surrounded by several adjacent molecules, forming regular geometric configurations. However, the regularity can only be maintained within several molecules, and the geometric configurations in different molecular clusters can be different. The geometric configurations can also fluctuate over time due to thermal motions. Such dynamic aggregation properties enable fluids to exhibit unique behaviors, making them invaluable for various opto-electronic applications, such as liquid crystal and RTP materials. Liquid crystal displays (LCDs), an early flat-screen technology, utilize the directional flow and alignment of liquid crystalline molecules to manipulate light under an electric field. This alignment controls light transmission, enabling image formation when combined with color filters and a backlight. Recent breakthroughs in fluid-based RTP materials, such as malic-acid-based systems,[163] have resolved challenges in achieving stable phosphorescence in fluid environments. These malic-acid-based materials demonstrate that supramolecular assembly in fluid state can mitigate these effects, opening pathways for in vivo applications and other advanced opto-electronic functionalities.

In conclusion, the versatility of molecular aggregates is reflected in their broad applicability across opto-electronic fields. For instance, nanoparticles in biological imaging and healthcare improve diagnostic accuracy and therapeutic outcomes. Thin films in photovoltaics and OLEDs enhance charge transport and exciton management. Crystalline materials provide insights into structure–property relationships, guiding the design of complex multifunctional devices. From organic solar cells to anticounterfeiting technologies, the aggregate states of molecules – whether

nanoparticles, films, or crystals – play a decisive role in achieving desired performance. Atoms make up molecules through chemical bonds, and molecular aggregates are formed through weak intermolecular interactions. The fancy performances of these materials not only depend on the properties of the well-designed molecules with specific function groups or/and fixed energy level, but also on their molecular packing, orientation, and interactions. The MUSIC concept emphasizes the centrality of aggregation in molecular design, urging further exploration of these states to unlock new development in opto-electronics. By integrating advanced aggregation strategies with innovative molecular design, researchers can continue to push the boundaries of performance and application in this field.

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