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Perspective

Opportunities for Next-Generation Luminescent Materials through Artificial Intelligence

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ABSTRACT: Luminescent materials are continually sought for application in solid-state LED-based lighting and display applications. This has traditionally required extensive experimental effort. More recently, the employment of data-driven approaches in materials science has provided an alternative avenue to accelerate the discovery and development of luminescent materials. In this Perspective, we give an overview of luminescent materials used for lighting and display applications with a specific focus on inorganic phosphors, quantum dots, and organic light-emitting diodes. We discuss recent progress using data-driven approaches to discover new compounds, predict optical properties, and optimize synthesis, among other topics for each type of material. We then highlight future research directions focusing on using artificial intelligence (AI) to advance these fields and address some cross-cutting challenges limiting the current application of AI techniques in luminescence-related research.

The "discovery" of naturally occurring luminescent inorganic materials transpired 400 years ago when Vincentinus Casciarolo of Bologna fired Bolognian stone, $BaSO_4$, in a charcoal oven. The carbothermal reduction of the barium sulfate mineral resulted in BaS, which emitted red light after exposure to sunlight.[1] Although the origin of the light was unknown, this observation spurred the search for other minerals that produced a similar response. The group of resulting luminescent compounds were termed phosphors after the Greek word for "light bearer." Yet, not surprisingly, the application of these fascinating materials goes back at least 1000 years when the Japanese reportedly used similar substances as paints.

The modern use of phosphors began in the last ∼120 years with ZnS-type compounds playing a central role in the original development of cathode-ray tubes.[2] This early research also identified numerous chalcogenide-based phosphors, recognized the importance of incorporating heavy atoms in the phosphor that act as the luminescence center, and established new spectroscopic techniques to characterize the optical properties of these materials.[3−6] The investigation of inorganic phosphors has accelerated dramatically in the last 70 years, driven not only by the advances in instrumentation and a desire to understand the physics and chemistry of luminescent materials but also by the promise luminescent materials show in an array of applications. Indeed, phosphors were initially used in television tubes, fluorescent lamps, electroluminescent displays, and X-ray screens. They have recently found an even more important application in solid-state, light-emitting diode (LED) based lighting.[7,8] Combining phosphors with blue or near-UV LED-chips in a single package results in the energy-efficient generation of white light. This pivotal breakthrough allowed the creation of light bulbs with dramatically lower energy consumption, environmentally benign components, and extended operating lifetimes.[9] The fundamental knowledge about phosphor materials generated by these early researchers has also allowed modern display technologies to replace old-fashioned cathode-ray screens with flat-panel LED/phosphor displays. The extensive use of solid-state luminescent materials has been made possible because the current generation of inorganic phosphors fulfills most of the application requirements: they have a range of excitation and emission wavelengths and bandwidths, high quantum efficiencies, outstanding chemical and thermal stabilities, and low manufacturing costs.[10−12] Nevertheless, there is still a need for new luminescent materials (inorganic and organic) with improved optical properties to meet the various needs of future applications.

The development of new luminescent materials for lighting and display applications still largely relies on simple design rules, systematic materials synthesis, or serendipitous discovery.[13] Today, there is an extensive understanding of optical physics that allows researchers to tune and optimize a material.[10,11,14] However, new luminescent materials must be discovered first. Researchers have, therefore, turned toward data-driven methods capable of accelerating the discovery process.[13,15−19] This

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approach extracts knowledge from past experiences by recognizing patterns and relationships in data. The data-driven materials discovery process typically follows the same basic workflow:[20] (i) construction of a data set that contains the known luminescent materials and their corresponding target properties; (ii) generation of descriptors to represent the materials, such as stoichiometric variables and structural information; (iii) recognition of connections and correlations between properties and descriptors; (iv) determination of screening spaces; (v) identification of materials with desired luminescent properties; and (vi) synthesis and characterization of the suggested compounds. The discussion herein focuses on the remarkable progress and outstanding success that data-centric methods have made recently with a specific focus on inorganic phosphors before expanding to some of the most promising new technologies, namely, quantum dots and organic light-emitting diodes. This Perspective suggests promising research directions with an eye on not only basic data-driven methods but also advancing toward using more complex artificial intelligence (AI) approaches for accelerating the discovery of luminescent materials.

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The current generation of inorganic phosphors used in most lighting and display applications converts a near-ultraviolet (n-UV) or blue LED chip emission into a broad spectrum of white light or specific colors for display applications.[7] Inorganic phosphors are composed of an insulating inorganic host compound, such as oxides, (oxy-)nitrides, or (oxy-)halide, and a luminescent center, such as a rare-earth ion or transition metal ion.[11] The host's coordination environment dramatically influences the orbital energies of the luminescent center, thereby controlling the electronic transitions. For example, in the rare-earth ions, changes in the crystal field splitting influence the optical properties by modulating the 4f ↔ 5d separation. The bonding between the luminescent center and the phosphor host also affects the phosphor's efficiency and the temperature-dependent optical properties.[7] The search for high-performance phosphors requires finding the optimum phosphor host and luminescent center pair. This is an intrinsically high-dimensional problem, but it may be possible to achieve the best combination of optical properties for a given application if successful.

Numerous data-driven research projects have already been carried out to find new phosphors with ideal optical properties. For example, studying the band structure of several well-known narrow-band red-emitting Eu^{2+}-doped nitride phosphors, Wang *et al.* found that the narrow bandwidth is characterized by a large splitting between the two highest Eu^{2+} $4f^7$ bands, as shown in Figure 1a.[21] They then incorporated this descriptor in the screening process and successfully identified five potential narrow-band red phosphors. Lai *et al.* correlated the crystal structure with optical properties in $A_3BSi_2O_7:Eu^{2+}$ (A = alkali metal ions; B = rare-earth metal ions) through a regression model to predict the emission wavelengths of Eu^{2+}-doped phosphors (Figure 1b).[18] Data-driven methods have also been combined with density functional theory (DFT) to guide the design and discovery of phosphors. This process follows a general workflow with the first criterion being that a phosphor must satisfy thermodynamic phase stability. This is described as the energy above the linear combination of energetically stable phases on the phase diagram (E_{hull}). The host band gap can then be used to narrow down the screening space as rare-earth substituted phosphors require compounds with a large electronic band gap. Moreover, the presence of a large band gap and rigid crystal structure tend to give rise to highly efficient and thermally stable materials. Beyond those general criteria, other factors also need to be taken into account in the search for specific phosphors. For example, in the search for broadband emission phosphors, multiple luminescent centers in the crystal structure are desired, which could be achieved via finding a host with diverse local environments for activators. This idea was applied to discover the full-visible-spectrum phosphor, $Sr_2AlSi_2O_6N:Eu^{2+}$, using a data-driven structure prediction model and data mining high-throughput DFT calculations, as shown in Figure 1c.[17] The green-emitting $Sr_2LiAlO_4:Eu^{2+}$ and blue-emitting $Sr_2LiAlO_4:Ce^{3+}$ were similarly discovered by data mining unexplored chemistries and applying DFT screening (Figure 2a−d).[22]

More recently, machine learning algorithms were employed to build mathematical functions between various phosphor data, known as "training data", and the desired output. An optimal function will generate the correct prediction with the given inputs that are not part of the training data. For example, a machine learning regression model capable of predicting the thermal quenching behavior was constructed based on the experimentally measured temperature dependence of more than 100 Eu^{3+}-doped phosphors.[23] These results provided a model capable of screening large databases of phosphor host materials and predicting the Eu^{3+}-substituted material's thermal response *a priori*. A tree-based ensemble learning model was also constructed to predict the 5d level centroid shift for Ce^{3+}-doped phosphors.[24] This model was also used to predict the position of the luminescent center's 5d-orbitals without any prior knowledge or expensive quantum mechanical calculations.

In some of the cases, explicitly predicting the optical properties is not straightforward. This is particularly true for problems where internal and external factors greatly influence the target properties. For example, a phosphor's quantum efficiency (QE) is highly influenced by the luminescence center substitution concentration, the measurement temperature, the synthesis route, and the postprocessing steps. Most of this information is not usually known for new materials. Therefore, in the search for highly efficient phosphors, explicitly predicting the QE is not necessarily possible. As an alternative, our group employed Debye temperature (Θ_D) as a proxy for QE, which is an intrinsic property that can be determined from the information available in high-throughput DFT computational databases.[16] After a machine learning model based on a combination of compositional and structural descriptors was built, the Θ_D values of more than 2000 compounds were readily predicted. Screening this data set for compounds that maximize the materials Θ_D and the electronic band gap revealed a novel, highly efficient blue-emitting $NaBaB_9O_{15}:Eu^{2+}$ (Figure 3a−c).

Emissive quantum dots (QDs) have gained significant popularity as another class of luminescent inorganic materials.

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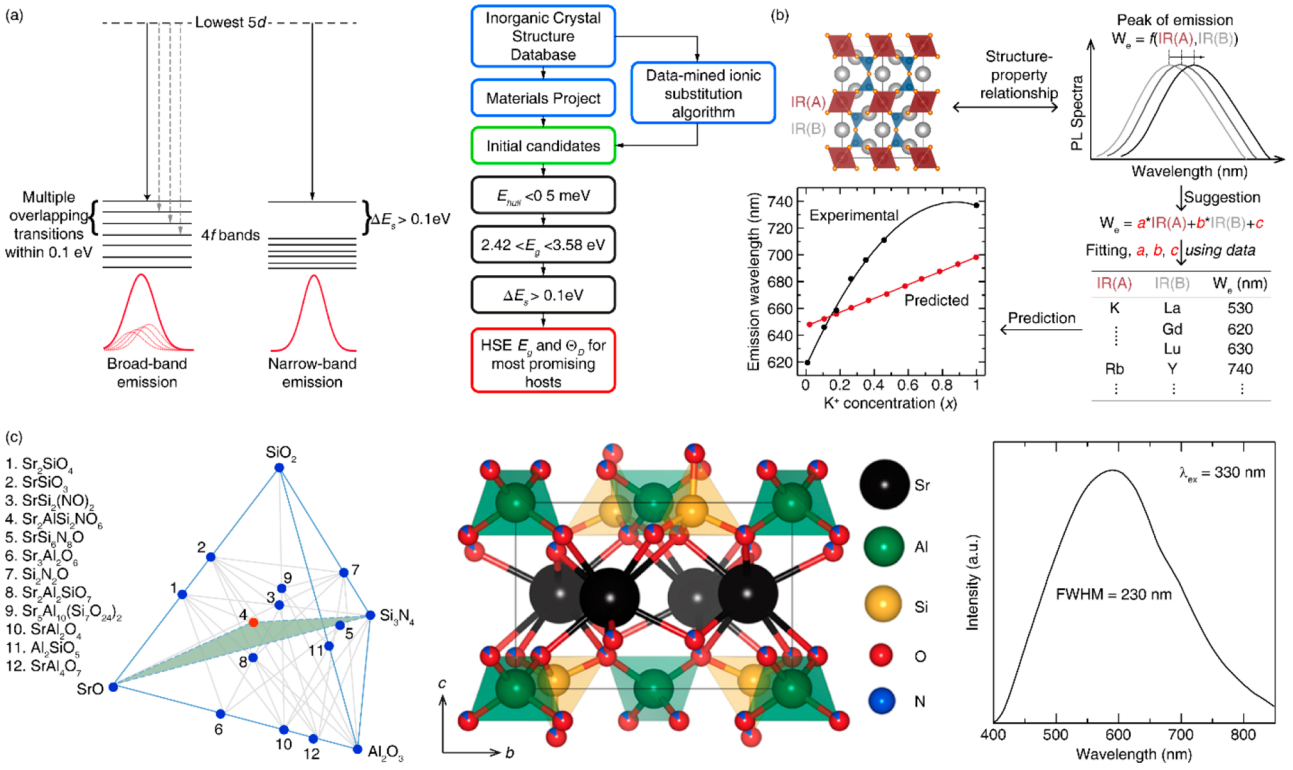

Figure 1. (a) Relationship between emission bandwidth and Eu^{2+} 4f band levels and flowchart showing high-throughput screening procedure for narrow-band red-emitting phosphor hosts. Reproduced with permission from ref 21. Copyright 2016 American Chemical Society. (b) Flowchart showing regression analysis steps on emission peak wavelengths of $A_3BSi_2O_7:Eu^{2+}$ system. Reproduced with permission from ref 18. Copyright 2020 American Chemical Society. (c) Calculated 0 K $SrO-SiO_2-Si_3N_4-Al_2O_3$ three-dimensional phase diagram, the energetically favorable crystal structure of $Sr_2AlSi_2O_6N$, and emission spectrum of $Sr_2AlSi_2O_6N:Eu^{2+}$. Reproduced with permission from ref 17. Copyright 2019 American Chemical Society.

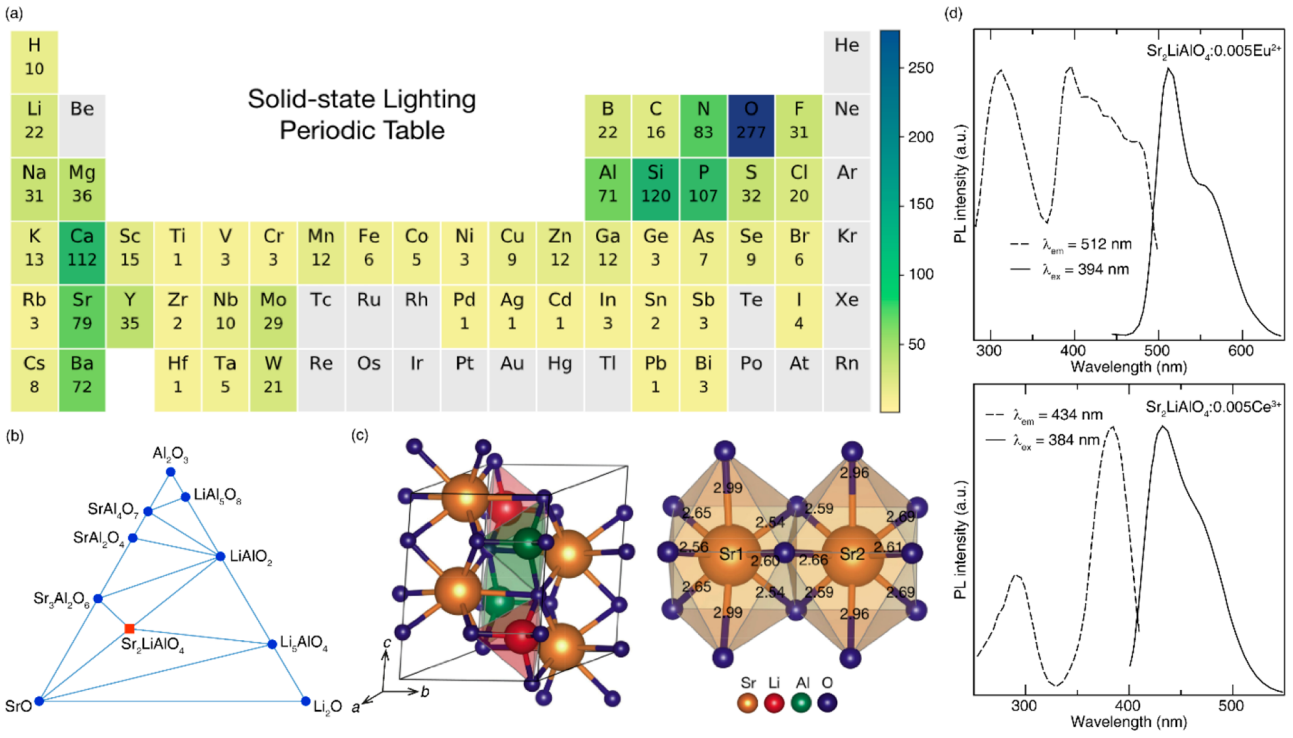

Figure 2. (a) Frequency at which each element appears in compounds having the word "phosphor" in the publication title in the 2017 version of Inorganic Crystal Structure Database (ICSD). (b) Calculated 0 K $SrO-Li_2O-Al_2O_3$ phase diagram. (c) Crystal structure of Sr_2LiAlO_4 and two symmetrically distinct Sr sites. (d) Excitation and emission spectra of $Sr_2LiAlO_4:Eu^{2+}$ and $Sr_2LiAlO_4:Ce^{3+}$. Reproduced with permission from ref 22. Copyright 2018 Elsevier Inc.

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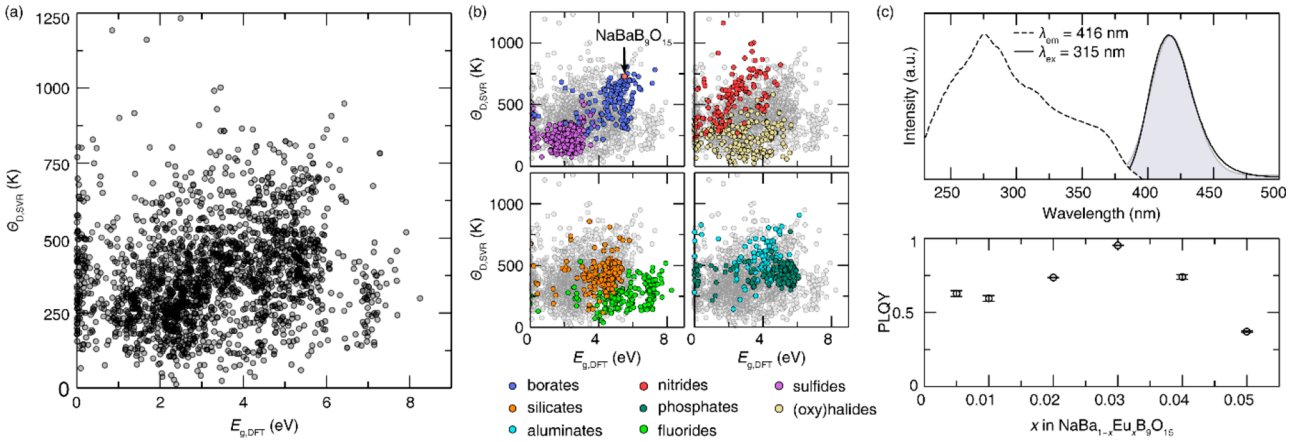

Figure 3. (a) Machine learning-predicted Debye temperature ($\Theta_{D,SVR}$) against the density functional theory calculated band gap ($E_{g,DFT}$) for 2071 inorganic compounds. (b) $\Theta_{D,SVR}$ against $E_{g,DFT}$ for classes of common phosphor host. (c) Excitation and emission spectra of $NaBa_{0.97}Eu_{0.03}B_9O_{15}$ and photoluminescence quantum yield (PLQY) of $NaBaB_9O_{15}$ substituted with varying Eu^{2+} concentrations. Reproduced with permission from ref 16. Copyright 2018 Springer Nature.

QDs are an alternative to ceramic, inorganic phosphors. They have shown tremendous growth because of advantages such as their small (nanoscale) size, easily tunable emission wavelength, narrow emission peak, high quantum efficiency, and short luminescence lifetime.[12,25−28] The most notable application currently is in LED backlight (display) technologies. The efficient and ultranarrow emission peak has allowed the red QDs, and in some cases green QDs, to replace the phosphor and color filters in the backlight of traditional LED-based displays. The resulting display covers a wider color gamut at a lower cost.[29,30] More recently, QDs have also been incorporated in on-chip LED-based general white lighting packages with excellent efficiency and outstanding color rendering. Advances in the chemical stability of the QDs through improved coating strategies, along with other modifications, have made these advances possible.

The most studied QD material is perhaps the Cd-based II−VI semiconductor like CdSe. This compound has been used in combination with other QDs for applications such as liquid crystal display (LCD) backlights and full-color μLED displays.[31,32] However, the toxicity of Cd notoriously limits its development and hinders commercialization. Alternatively, the III−V QDs, such as InP, are Cd-free, but they show slightly lower quantum efficiencies and a broader emission band.[33] The ABX_3 perovskite QD materials, on the other hand, have recently attracted interest because of their outstanding optical properties. They show an even narrower emission band than the II−VI and III−V QDs. This allows the color gamut to cover beyond the National Television System Committee (NTSC) specification by as much as 128% and cover the ITU-R color space recommendation BT.2020 (Rec. 2020) by 85%.[34,35] However, the perovskite QDs also tend to contain heavy metals like lead, which pose environmental and health concerns similar to those of Cd. Therefore, developing more environmentally benign heavy-metal-free QDs with desired optical properties has been a significant research focus.

Researchers have started to address this challenge by applying data science to optimize the manufacturing and processing of new quantum dot materials. The rapid parameter screening provided by data-driven automation can accelerate the product's optimization with minimal laboratory effort. This substantially drives down labor and material costs. For example, an automated microfluidic platform combined with *in situ* photoluminescence monitoring allowed researchers to map the acid:ligand ratio and reaction temperature space. The result was an approach capable of identifying the reaction conditions that yielded quantum-confined nanostructures with high brightness and narrow emission peaks.[36] Regonia *et al.* also developed an artificial neural network (ANN) model to predict the optical band gap of ZnO QDs based on the reaction time and temperature.[37] Moreover, ANNs have shown great potential in the optimization of QD synthesis. For example, Bezinge *et al.* reported a self-optimizing algorithm that automatically identifies the reagent concentrations that yield perovskite nanocrystals with a specific emission peak wavelength (Figure 4a,b).[38] Likewise, as shown in Figure 4c−e, Epps *et al.* integrated an ensemble neural-network-based experiment selection with a high-efficiency autonomous flow reactor to synthesize perovskite QDs with maximum photoluminescence QE and minimum full width at half-maximum (fwhm).[39] Bayesian optimization has also been implemented by Voznyy *et al.* to improve the monodispersity of PbS QDs.[40] Finally, machine learning has been used to investigate the operation mechanisms in QD-based devices. For example, a random forest regression model showed that the current limiting factor of red-emissive QD LEDs is the hole injection barrier from the hole transport layer to the QD layer. This finding suggested that reducing the hole injection barrier can enhance efficiency.[41]

Beyond inorganic compounds, organic light-emitting diodes (OLEDs) have also received significant attention, especially in the area of flat-panel displays. These luminescent organic compounds form a flexible and bendable thin film that acts as an emissive electroluminescent layer that emits light in response to an electric current. OLEDs are also diffuse direct light sources, which allow them to be safely placed very close to the illuminated object without risking damage.[42] The flexibility in shape and diffuse light emission also offers outstanding opportunities for lighting designs not available with traditional lighting technologies. For instance, compared to LED displays, the electroluminescence of the OLED means that it can work without a backlight, which makes the product thinner and lighter.[43] Moreover, OLED screens can achieve higher color contrast and response time at lower power consumption than traditional flat panel displays.[43] However, OLED screens are

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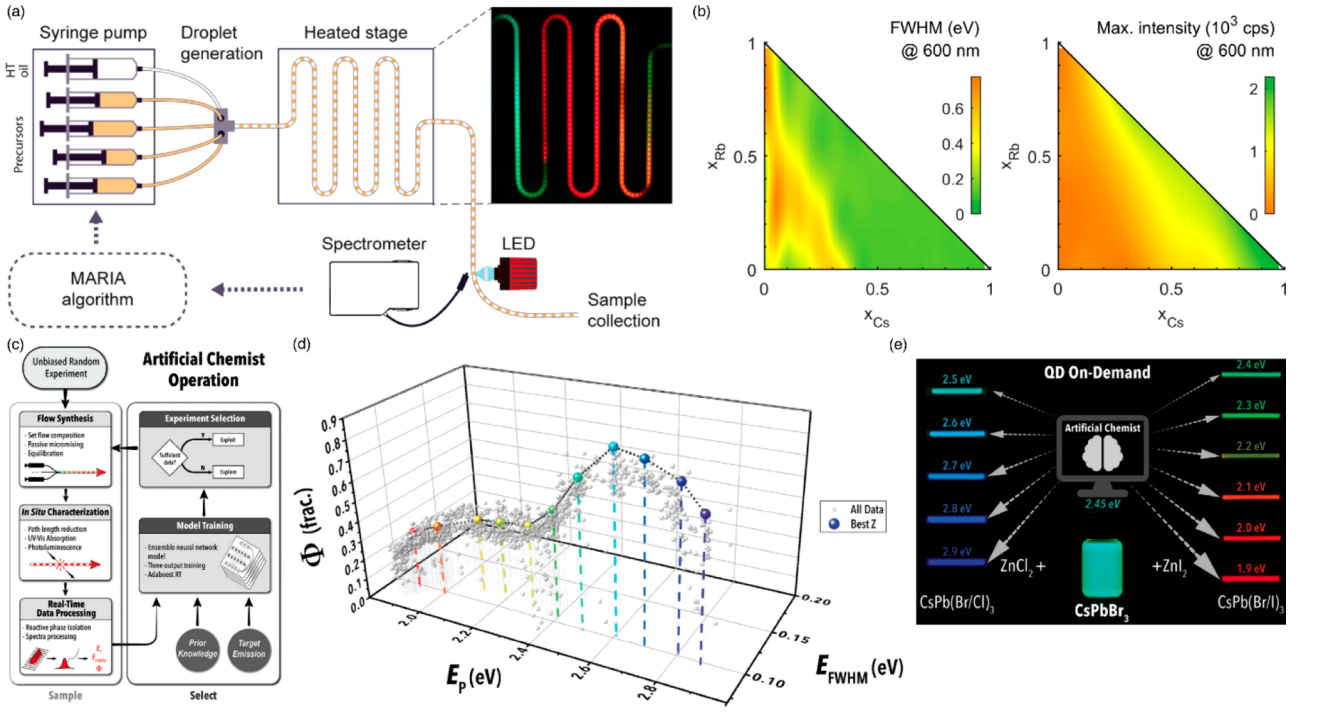

Figure 4. (a) Schematic of the experimental setup for the synthesis of (Cs/FA)Pb(Br/I)3 (FA = formamidinium) nanocrystals. (b) Predicted fwhm and maximum intensity of nanocrystals under the reaction conditions predicted by the algorithms. Reproduced with permission from ref 38. Copyright 2018 American Chemical Society. (c) Flow diagram detailing the developed smart modular fluidic microprocessor for autonomous synthetic path discovery and optimization of colloidal QDs. (d) Photoluminescence quantum yield (Φ) as a function of peak emission energy (E_p) and emission line width (E_{fwhm}) for all 1400 in-flow collected data points with the 11 optimal formulations highlighted. (e) Illustration of the UV-illuminated starting QDs and final halide-exchanged synthesized in flow for all 11 target emission colors. Reproduced with permission from ref 39. Copyright 2020 Wiley.

currently appropriate for small to medium-sized devices, such as television, computer monitors, and portable systems such as smartphones. Large screen applications still dominantly rely on the phosphor/LED scheme. The factors that prevent OLED from being applied currently to ultralarge screens are the limited lifetime of the organic materials, color imbalance over time, high manufacturing cost, and high power consumption.[44]

OLED material technologies are primarily based on small molecules and polymers. One of the most commonly used small molecules are organometallic chelates, *e.g.*, tris(8-hydroxyquinolato)aluminum(III) (Alq_3), which was first developed by Tang *et al.* in 1987.[45] The production of small molecule devices usually involves thermal evaporation and vacuum deposition of the small molecules to deposit a thin film, which enables flexibility in layer design. Contrary to small molecules, polymers are processed in solution and spin-coated to form thin films. This method is more suitable for forming large-area films. The landmark discovery of poly(phenylenevinylene) (PPV) in 1990 led to the development of solution-processable OLED materials.[46] With OLEDs, light is generated from the organic layers by the formation of excitons through electron−hole recombination and emission of radiation. Enhancing the possibility of exciton energy leading to photon emission and controlling the flow of electrons and holes to the recombination region are the keys to increasing light production. Therefore, besides basic optical requirements, such as high quantum efficiency and outstanding color purity, other OLED material requirements include good film-forming properties and good carrier mobility. Although there are still significant challenges facing these materials, they are quickly becoming ubiquitous in the marketplace, at least for display applications.

Data-driven methods have only recently been applied to search for new OLED materials and optimize the fabrication process. For example, Gómez-Bombarelli *et al.* integrated a machine learning model with computational quantum chemistry to screen over a million candidates in the search for an efficient blue-emitting OLED (Figure 5a−f).[47] Choi *et al.* employed supervised learning techniques to model the power consumption through the RGB value of each pixel of the OLED display in mobile devices.[48] Janai *et al.* took a random forest algorithm to extract the complex correlation between blue OLED device efficiency and device parameters such as triplet energy, frontier molecular orbital energy levels, and layer thickness, finding that the triplet energy of the electron transport layer is critical to OLED efficiency.[49] There is clearly ample opportunity for applying data-driven methods to develop OLED materials.

Researchers have traditionally relied on experimental work for the exploration of luminescent materials. Employing ideas rooted in data science, specifically artificial intelligence methods, has only recently been employed to advance the field. There are plentiful research opportunities available for exploration. For example, near-IR and laser phosphors have also shown great potential in a range of applications. NIR-phosphor-converted LEDs enable fast and convenient diagnostics and food analysis when coupled to a smartphone. Laser-excited phosphors also offer a viable option for ultrahigh-power lighting.[50,51] Although, these devices are not been widely implemented yet, the assistance of AI may significantly accelerate the development of these materials and spur their use. Moreover, the temperature

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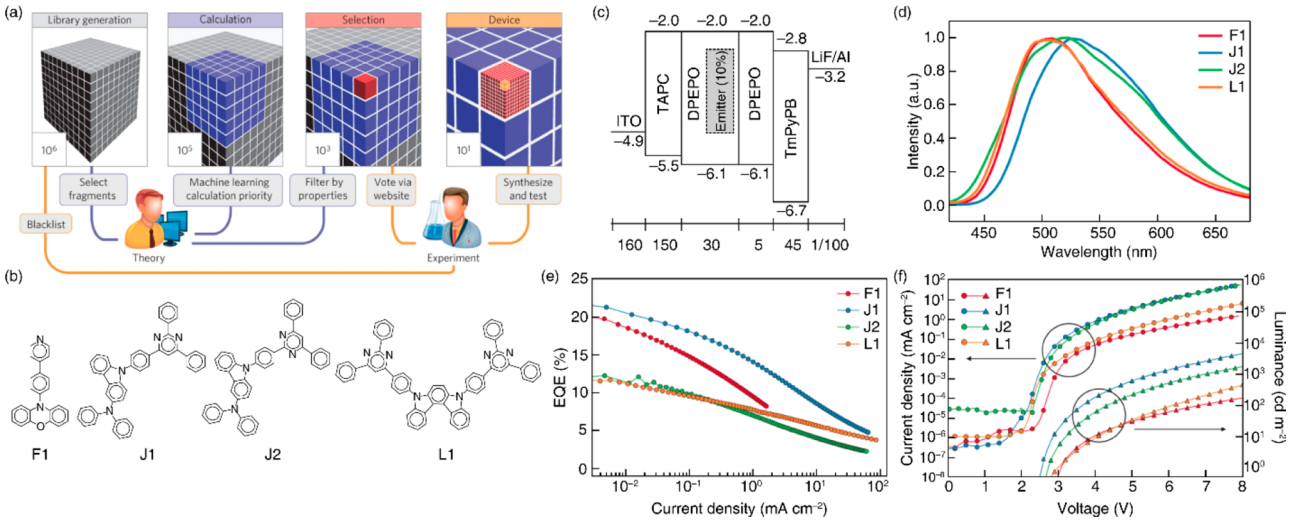

Figure 5. (a) Diagram of the collaborative discovery approach. (b) Promising molecules that were synthesized and tested. (c) Device structure and energy band diagram of lead candidates. (d) Electroluminescence spectra. (e) External quantum efficiency (EQE) as a function of current density. (f) Current density and luminance as a function of the applied voltage. Reproduced with permission from ref 47. Copyright 2016 Springer Nature.

near the backlight of an LCD screen can be around 100 °C. Above this temperature, the luminescent material loses efficiency and becomes less bright.[52] Synthesizing luminescent materials with high quantum efficiency and good thermal stability is critical to improve the quality of these displays. Debye temperature and band gap are usually used as proxies for QE and thermal stability.[53−55] However, there are many examples where these proxies break down. It is possible to instead use AI to directly predict QE and thermal stability for the specific materials by training a machine learning algorithm to directly predict the temperature-dependent luminescence. These models could ideally be used for materials discovery by screening composition spaces that maximize these models. AI also has the potential to identify promising materials when combined with high-throughput synthesis methods. Moreover, synthesis and postprocessing optimization of luminescent materials, such as compositional engineering, surface engineering, and device encapsulation, have proven vital for enhancing the lighting and displaying performance. Machine learning can unify all of the synthesis and postprocessing factors to establish a comprehensive link between the specific optical properties and the preparation conditions.

Moreover, a significant amount of the previous data-driven methods to discover luminescent materials are designed to predict optical properties in a simple, single numerical value, *e.g.*, fwhm or emission peak wavelength. This process results in the loss of a large amount of information embedded in the spectroscopic data, such as the peak shape. One possible future direction is to learn from the spectroscopic data directly. One recent publication showed it was possible to predict the UV−visible absorption spectra for materials through a series of machine learning models.[56] It is conceivable that a similar approach could be developed for photoluminescent data (excitation/emission curves). In another study, a machine learning model based on time-resolved photoluminescence spectroscopy was constructed to determine the distribution of decay rates in $CsPbBr_3$ perovskite nanocrystals without any *a priori* assumptions on the underlying physics governing the photon emission. This model could provide insight into the physical mechanisms of luminescence.[57]

Although substantial efforts have been made with data-driven and AI methods in developing luminescent materials, significant challenges remain. The most crucial component in data-driven and AI methods is the quality of data available. Maximizing data accessibility and finding appropriate applications of the data are keys to constructing robust models. This is just one among many other cross-cutting challenges that must also be addressed for AI to make truly transformative changes in the discovery of luminescent materials.

The most crucial component in data-driven and AI methods is the quality of data available. Maximizing data accessibility and finding appropriate applications of the data are keys to constructing robust models.

Data Organization. The field of materials informatics is emerging in parallel with the growth of materials databases. Many of these independent data sources are quite general, and they usually provide only basic knowledge about compounds, *e.g.*, structures or fundamental physical and chemical properties. Although there are many new publications on luminescent materials coming out daily, accurately gathering the desired data and turning this into structured information is one of the biggest hindrances in utilizing data-driven methods. It is somewhat ironic that we have access to more data than ever, yet we continuously face data shortages. Creating a platform or database to organize the luminescent data systematically would significantly increase data accessibility. Access to such a database will undoubtedly benefit researchers who are investigating luminescent materials.

Learning from Small Data Sets. Even if a database of optical properties were created, these data sets remain sparse and barely large enough to construct informative and statistically robust models. This has been one of the pervasive challenges in this burgeoning field. Researchers have recently shown it is possible to efficiently expand data sets through automation. Text-mining

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algorithms, for example, have proven to be effective at extracting data from the materials science and chemistry literature, although it requires the creation of a reliable, unique identifier for each entity, *i.e.*, matching synonymous terms.[58−60] Moreover, for problems where data are intrinsically scarce, such as the crystal field splitting, employing more advanced data-driven techniques might help. For example, adaptive design, also called active learning, is a powerful method for constructing reliable models with small data sets. This approach accumulates new knowledge iteratively by introducing feedback from experiments into the training set in each loop. The application of adaptive design has expanded into materials science recently and obtained some success in materials discovery.[61,62] In addition, transfer learning can pass the knowledge gained from a previous model to solve a new but related problem. For example, Dorenbos *et al.* showed the existence of relationships between inorganic phosphors doped with different rare-earth ions. Knowledge gained while training a Ce^{3+}-doped phosphor model could apply to other rare-earth substituted phosphors, such as Pr^{3+} and Nd^{3+}.

Efficient Data Representation. When attempting to apply data-driven approaches, each material needs to be represented by a set of finger-printed features after the training data sets have been created. In most cases, features are numerical and in the form of tensors, either vectors or matrices. Data representation can be described as a "connection" between input and output data. In general, a more suitable data representation leads to a better model, which can effectively capture the hidden structure of input and output data to give a more accurate prediction. Unfortunately, there are no unified feature sets that are suitable for machine learning studies. Personalized features are often required in each specific task to maximize the model's accuracy. Although massive research effort has gone into encoding complex materials data, developing features more specifically for luminescent materials is still critical. In the case of inorganic (ceramic) phosphors, features are mainly based on property-related compositions and average crystal structures. Although these features capture a diverse range of physical and chemical properties of the compounds, models to predict optical properties, such as excitation wavelength, sometimes suffer from low accuracy because of difficulties representing the luminescent center's local environment. This local structure information and distortions in the coordination environment are more influential than the average structure for determining the orbital interactions, crystal field splitting, and optical properties. However, this information often remains unknown. It can even be difficult to assign the coordination numbers of the luminescent center. For example, the Eu^{2+} in $CaAlSiN_3$:Eu^{2+} phosphor was considered to be four- and five-coordinated in two different works.[63,64] Therefore, algorithms to represent the local geometries are essential to promote this field to the next level.

This Perspective presented a brief overview of the latest progress made by data-driven methods in the discovery and development of luminescent materials, specifically, inorganic phosphors, QDs, and OLEDs with applications in lighting and display technologies. Although remarkable progress and exciting discoveries have been made recently using data science, an appreciable number of opportunities and challenges remain. Some future research directions are proposed here, and some general difficulties limiting the application of AI techniques in luminescence-related research are identified. Addressing all of these challenges will undoubtedly lead to a new generation of luminescent materials.

Although remarkable progress and exciting discoveries have been made recently using data science, an appreciable number of opportunities and challenges remain.

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Notes

The authors declare no competing financial interest.

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