The Recent Development of Blue LED

Jingqian He[1, †], Muyan Ma[2, †], Zhenzhong Tang[3, *, †], Junchuan Wu[4, †]

[1]Department of Physics, Grinnell College, Iowa, USA

[2]Department of Science, Beijing University of Technology, Beijing, China

[3]Department of Physics, Southern University of Science and Technology, Shenzhen, China

[4]Department of Physics, The University of Melbourne, Melbourne Victoria, Australia

*Corresponding author: 11910525@mail.sustech.edu.cn

[†]These authors contributed equally to this work.

Abstract: LED is a very influential light source for its high brightness, good color display and ability to modify the spectra. The performances of red LEDs and green LEDs have been greatly improved but the blue LEDs' performance is still left behind. In this paper, researchers investigated the frontier research in recent years, including the progress and possible direction in conventional inorganic GaN blue LEDs, quantum dot blue LEDs, organic blue LED, and perovskite blue LED. The basic working principles of each direction are introduced first and the related characteristics of the materials and the luminance mechanisms are discussed. Then the authors analyze the key influencing factors in each of the above directions and compare the development status, LED performance parameters the stability, luminance efficiency and related advantages and disadvantages. Besides, brief history of the development will be introduced to get a full view of the changes in research idea in the past years. In each direction, a discussion is made with the focus on applications, further challenges, and the possible improvements and some suggestions and expectations of the blue LEDs are put forward.

Keywords: Blue LED, Organic LED, Quantum Dot, Perovskite.

1. Introduction

In today's technology, semiconductor light-emitting diodes (LEDs) are a crucial element. They bring great convenience to people's daily life and the improvement of the science. They can be used as glowing indicators in computer, car brake lights and traffic lights [1]. There are also an important link between electronics and photonics technology, and have shown great value and potential in recent decades and in the future.

A solid-state device called a light-emitting diode transforms electrical energy into light energy. Its basic structural components are a PN chip, an electrode, and an optical system. A photoelectric conversion process underpins the fundamental operation of LEDs [2]. The electrons in the N region of the LED will be pushed to the P region when forward voltage is applied. In the P region, holes are pushed to the N region and then emit energy in the form of photons.

Incandescent bulb is a mixture of various wavelengths of light generated by heat, while fluorescent lamps are the first to produce the highest energy ultraviolet, and then rely on ultraviolet light to excite the phosphor, producing a variety of lower energy visible light. LED single red light is not available white light, can only be used to be monochrome light source or signal lights on the instrument. Painting a certain percentage of phosphor on the blue LED can help excite the low energy yellow-green light based on the high energy blue light, and then white light can be produced when mixed with red light. So before the birth of blue LED, neither the display nor lighting can be achieved with LED, and only with the invention of blue LED these can be widely used.

Red LED was invented for the first time in 1950s. By the 1960s, green LED was yielded [3]. However, the technology of fabricating blue LED was a huge problem. Until 1970s, there was significant advancement in processes like hydride vapor phase epitaxy which is also named the HVPE, liquid phase epitaxy which is also named the LPE, and later molecular beam epitaxy which is also

named the MBE and metal organic chemical vapor deposition which is also named the MOCVD. GaN and InGaN material advancements paired with device structural modification, such as the inclusion of an AlGaN layer to reduce leakage current, produced blue LEDs with improved performance that, in 1994, attained an astounding blue light luminous intensity more than 1 cd. Therefore, commercialized blue LED became possible. In recent decades, the technology of organic LED, quantum dot, PVSK have experienced rapid development. For organic LED, It is a developing lighting technique, particularly for small displays like cell phones and other devices. Compared to traditional technology, it can be made lighter and thinner. Although red and green LED are already quite successful, blue LED still faces great challenges because of its short operation lifetime. In some inspiring and creative research in recent years, significant advancements were achieved in our knowledge of physical mechanism and technology approaches underlying electroluminescent devices that use organic active materials. While there have been many improvements, the research that has increased quantum efficiency and operating lifespan to levels that call for serious consideration of such devices for a variety of applications has seen the most significant gains. In order to solve the problem, another new technology, Deep blue LEDs could be created from colloidal quantum dots, which are normally made of heavy metals like lead and cadmium. Transistors, photovoltaics, and other components such as those listed above come with deep blue LEDs [4]. Quantum dots are small groups of semiconductor particles. They have great advantages in the area such as stability, high efficiency and the potential to commercialize. For the PVSK blue LED, many researchers are working to improve stability and reduce toxicity. It demonstrates the opportunities to reduce manufacturing costs and the promise of flexible substrates for roll-to-roll manufacturing, so that allowing, for instance wearable devices. In 2014, Tan's reported an increased external quantum efficiency tremendously by PVSK technology. Finally, it is equivalent to other LED technologies. It achieves great success in the application of Peled in the field of commercial application of displays and lighting.

This article, aims at reporting on blue LED from the perspective of conventional inorganic LED, OLED, QLED, PeLED, and compare their advantages and disadvantages in the areas such as performance, cost, practicality, etc. Their further research prospects are also investigated.

2. Conventional inorganic blue LED (GaN-based)

For the blue LED, researchers found 3 possible materials with appropriate energy band gap (ZnSe, 6H-SiC, GaN). However, SiC has an indirect band gap and ZnSe has very low stability for its low bonding energy and difficulty to make a good crystal. Finally, they found the possibility of blue LED in GaN with a direct band gap in 1971[5]. Then Akasaki, Amano, & Nakamura made high brightness LED with high quality single crystal of GaN and solve the crystal growing problem by vapor-phase reaction. In 1969, Maruska and Tietjen used the hydride vapor-phase epitaxy (also be known as the HVPE), so that a single crystal grew GaN films on substrates made of sapphire. The HVPE method uses the reaction of ammonia (NH3) with gallium chloride at temperatures of roughly 1300 K to crystallize GaN on a foreign substrate at ambient pressure. [6] In 1973 and 1975, GaN was made by Akasaki using hydride vapor-phase epitaxy (HVPE) and molecular beam epitaxy (MBE). MBE technique is an epitaxy method in which single crystals were made by deposition. It requires high vacuum in order to achieve the same impurity levels as other deposition methods. It is also the fundamental technique for the development of nanotechnologies. In 1978, Akasaki used HVPE to make a MIS (metal insulator-semiconductor) GaN blue LED with an efficiency about 0.12% but with low brightness under high voltage. Akasaki then found MBE will cause the lack of nitrogen and slow growth rate while HVPE cannot ensure the quality of crystals. In 1979, he finally decided on MOCVD[7]. In MOCVD, a reactor is often supplied with a non-reactive carrier gas together with ultrapure precursor gases. The precursors undergo pyrolysis when they get closer to the semiconductor wafer, and the products then soak onto the surface. A new epitaxial layer of the semiconductor crystal lattice is created as a result of the surface reactivity of the precursor material. Compared to MBE and HVPE method, it does not have the reverse reaction. It also is much easier to

control growth rate and allow the impurity doping. In 1990, Nakamura could make a very thin GaN layer at a relatively high temperature. In 1989, Akasaki, Amano, Koide added LT-AlN buffer layer to enhance GaN crystal quality and improve the conductivity in the following research. Resulting from the high pressure that the dissociation of nitrogen needs at the development temperature of GaN (1000 °C), it can be challenging to directly prepare a bulk GaN single crystals. So, an alternative method is to make the heteroepitaxial growth on some substrates. [8,9] In 1994, Nakamura and colleagues used a double heterojunction with a specific designed structure of InGaN/AlGaN to obtain a quantum efficiency up to 2.7%. During the research process, the main subject, for blue led which is mainly just GaN[10]. In order to make high quality blue LED, we need to be able to produce high quality Gan films. Therefore, we change some important variables, such as the system of doping GaN, until we get expecting result and we analyze the result and compare with previous studies.

3. Blue OLED (organic LED)

An organic LED is a kind of LED with a film of organic compounds to serve as the emissive electroluminescent layer to emits light with an electric current. As a new developing technology, OLED is mostly used in small displaying equipments. Compared with traditional LED, OLED can be grown in thinner size, lighter and cheaper in commercial production. Researches on green and red OLEDs have already achieved great success, but the operational lifetime is still a struggling study obstacle for blue OLED which is crucial for the full-color display device production. The dissociation of molecules, flaws, and the emergence of radical species are the primary causes of the short lifetime. These processes are created in organic materials through a variety of uni- and bimolecular processes, including direct photolysis, exciton-exciton contact, and exciton-polaron interaction. The fundamental experiments are still needed for the massive production, but the novel developing techniques in industry have provided several possible solutions for the future success of blue OLED including phosphorescence (Ph), thermally activated delayed fluorescence (TADF), fluorescence (F), hybridized local, and charge transfer (HLCT) OLED[11,12].

3.1 Fluorescence blue OLED

An important technique used in fluorescence blue OLED (F-OLED) is called triplet–triplet annihilation (TTA). It takes a long time for phosphorescence to transition start from a triplet state, and ends to the ground state using the conventional method of light excitation, thus limiting internal quantum efficiency (IQE) to only 25%. In TTA process, through the triplet fusion, light is emitted by two triplets fusing back to a singlet, which can boost the IQE to 62.5%. The use of a twisted arrangement with a bisanthracene core can provide blue light emission, which reduces the conjugation. In order to prevent the bathochromatic effect, a weak electron donor (methoxy, OMe) and acceptor (cyano, CN) are also added to the end groups that need to be attached. While maintaining the deep-blue emission, the photoluminescence quantum yield is significantly improved by adding butoxyphenyl to the anthracene core [13].

3.2 Phosphorescence blue OLED

For the operating mechanism, 75% triplet and 25% singlet excitons are formed in electrical pumping and then by intersystem crossing, singlet excitons transform to the triplet state. The heavy metal ions, iridium are added to the organic materials to improve spin-orbit coupling, the lifetime of triplet exciton is lessened to microsecond level and facilitates radiative relaxation to ground state[6]. In order to produce blue light emission, the highest occupied molecular orbital must be deeper, one straightforward method is to introduce electron-withdrawing units into the phenyl ring of pyridine and one of the earliest example phosphors is bis[2-(4,6-difluorophenyl) pyridinato-*C*2,*N*] (picolinato)-iridium(iii) (FIrpic). Then, N-heterocyclic carbene (NHC) was developed to achieve deep blue phosphorescence OLED emission with a constrained full-width half-maximum (FWHM) [14,15].

3.3 TADF OLED

The process by which a chemical species in an excited, non-emitting state absorbs ambient heat energy to change states and release light is known as thermally activated delayed fluorescence, or TADF. An excited molecular species in the triplet state is the major player in the TADF process. Reverse intersystem crossing (RISC) and absorption of ambient thermal energy allow the triplet state to become a singlet state, which can subsequently deexcite to the ground state and release light. To create a TADF emitter, a suitable center bridge or dihedral angle must be constructed between the electron donor and acceptor. It is necessary to use weak electron acceptors like diphenylsulfone and strong electron donors like acridan to fabricate a blue emitter with a comparatively wide energy bandgap. Compared with Ph OLEDs, TADF OLEDs are more flexible for the industrial production since they are pure organic material without the needs of combining with heavy metal atoms. However, just like all other OLED, TADF OLEDs are also constrained by the limited lifetime[16].

3.4 HLCT OLED

A high PLQY is one of the most important criterion for luminous materials. Due to the low price, less perniciousness, and high efficiency, pure organic substances that can achieve a hybridized local and charge-transfer (HLCT) state are candidates for use in high-efficiency OLEDs. In a single molecule, a high-energy charge-transfer state forms and thus, at an excited state, the reverse intersystem crossing may occur to transform the triplet to a singlet. The light is emitted by the excited singlet relaxing to S1 with a high PLQY. To form HLCT molecules, a high level of CT is needed to combine the singlet and triplet. In order to obtain high efficiency, the PLQY of LE1 is required to be high. Meanwhile, to prevent triplet excitons from relaxing from Tn ($n > 1$) to the dark T1 state, LE3 should have a low energy[17].

4. Blue quantum dot LED

Quantum dots (QDs) are small group of semiconductor particles that has similar electronic states as single atoms. They have some huge advantages in high illumination efficiency, good color purity and stability, low cost, and ability to mass production and they have potential usage in many areas. The different size of QDs determine different band structures and then different color of light when QDs are excited. QDs LEDs have very high degree of freedom in device design for the easiness to control the color display. The performance parameter of blue QDs LEDs still needs to be improved especially when it is compared to the red and green ones. Traditional Cd based and Pb based QDs are toxic despite their high performance. The heavy-metal-free QDs have the peak of the EL emission larger than 480nm and have huge performance margin and obvious development potential, which still need better synthetic methods to make uniform-size-nanocrystals with fewer defects and more suitable structure of device to balance the injection of charge and control the transportation. After years of development, many potential blue QLEDs have been developed. Zn based and carbon QDs are the research directions with more progress in recent years.

4.1 Zn-based

The intrinsic toxicity of traditional Cd based QDs and Pb based QDs limits their further application and the heavy-metal-free QDs are ideal directions. Among the heavy-metal-free QDs, Zn based QDs are the ones with most progress. ZnSe has a direct band gap conform the wavelength of blue light (2.7eV) that confines both electrons and holes to the core region to ensure PLQY and stability. Many advanced QDs a based on ZnSe.

4.1.1 ZnSeS (GCS-QDs)

GCS-QDs is structure of ZnS/ZnSe core with majority of Se is located at the core and S is mainly distributed on the shell. Its emission range is 420-440 nm (blue light > 440 nm). A structure with continuous gradient shell is made to weaken the quantum confinement effect to make an emission of

445 nm and suppress the energy transfer between films and other non-radiative energy transfer processes to higher the PLQY. The narrow size distribution (<10%) reduces the FWHM of the PL spectrum to improve color purity. An EQE of 3.5% is made and the value of FWHM is made to 12 nm at 445 nm and 16 nm at 448 nm. It also has a PLQY of 78% and brightness of 3359 cd/m^2. The perfect lattice match also ensures the PLQY and stability (keeping 80% of the luminance after 60 hours under continuous ultraviolet rays with 420 nm and optical density = 1.0). Without the gradient shell, it would have an obvious decrease after 6 hours and down to 40% after 30 hours[18].

4.1.2 ZnTeSe QDs (C/S/S QDs)

C/S/S QDs have a structure of ZnTeSe/ZnSe/ZnS and 2 gradient chloride content emitting layers were constructed to improve hole transport to achieve much higher brightness, much higher efficiency, and much longer operational lifetime. Te doping changes QDs' emission wavelength to 457 nm and the PLQY can be improved by eliminating faults of attachment and stacking (which will easily generated when the core formed) and surface defects from passivation in the ZnSe structure with HF and ZnCl2. It makes an EQE of 20.2%, brightness of 88,900 cd/ m^2 and T50 of 15,850 h. HF can detach the ligand, higher PLQY to 70% and suppress the average decay time; $ZnCl_2$ can replace the bulky ligand and higher the PLQY to 77% and further increase tau to 77 ns. QLED using both HF and ZnCl2 exhibits a high PLQY of 93%[19].

4.1.3 $CuInS_2$/ZnS

Cu based QDs have the emission range from green to red. It can be changed to blue light by connecting the alloying Cu–In–S QDs with ZnS which has a greater band gap. High color rendering index (CRI) requires a wide emission spectrum, hence QDs with a restricted emission range are insufficient. The $CuInS_2$/ZnS core/shell quantum dots have a wide emission band and are non-toxic. The optical band gap will be wider if the reaction time for core creation is sped up and the emission peak redshifts from 680 nm to 780 nm, according to the CuInS2 QDs' absorption and PL spectra. After the ZnS shell is added, the blue shift will happen because the similar lattice constant makes the combination of $CuInS_2$ and ZnS have a smaller $CuInS_2$ core. By reducing the ratio of Cu to In, the light of $CuInS_2$/ZnS moves from red to blue and has a quantum yield (QY) of 67%. When the dots combine with polystyrene, the luminous efficacy can be made to 10.7 lm/W. However, the degradation will happen within 4 hours[20].

4.2 Carbon based

CDs have many surface function groups and the optical properties can be modified through changing the function groups. However, most CDs are highly excitation dependent for the surface defects and that limits the injection of the carriers.

Oleophilic CDs based LED was fabricated in 2013 with a QY of 60% and brightness up to 24 cd /m^2. In research in 2018, CD-LED demonstrated a brightness of 569.8 cd/m^2 and QY up to 41%. The concentration of CDs in PVK will affect the energy transfer from PVK to CDs, which in turn will affect the concentration of CDs in PVK. The blue peak in the EL spectra will shift from 400 nm (PVK) to 474 nm when the concentration rises (30%). (CDs). With even higher concentration (>30%), the brightness will decrease for the luminescence quenching from the long chain ligands of CDs. Therefore, the concentration is usually set to 30%. These findings suggest that oleophilic CDs are superior to hydrophilic CDs for high brightness and clean emission because PVK is only soluble in chlorobenzene and chloroform and because it is challenging to regulate the concentration of hydrophilic CDs. The CD's LUMO Energy levelis-3.4 eV and the offset from that of TPBi (-2.7 eV) is -0.7 eV, which makes it easier for the injections of electrons. Besides, the HOMO Energy level is -6.4 eV and the offset from TPBi (-4.7 eV) is -1.7 eV, which prevents the injections of electrons.[21]

Research in 2020 found that the functional groups containing oxygen will help to broaden the spectrum, so elimination of the oxygen-containing functional groups will help to make a narrow emission. Besides, additional surface amination steps are made to reduce defects and suppress the non-radiation ways to ensure high color purity deep blue CDs (HCP-DP-CDs) with a narrow

absorption peak at 395nm, emission peak at 433nm and FWHM of 35 nm, which is very small compared to the normal ones that FWHM longer than 80 nm. The parameters are PLQY of 70%±10%, maximum luminance of 5240cd/m^2, maximum EQE of 4%, and PL excitation peak at 394nm. Its stability can keep for about 6 months without obvious change. After working for 3 hours with initial luminance 1000 cd/m^2 and constant current density 40mA/cm^2, it will retain 50% of the luminance[22].

There are no mature methods to realize the mass production of LEDs that are treated in solutions and there are no excellent approaches on large-scale synthesis of colloidal quantum dots, which raises the expenditure of QD based emitting layer manufacturing. Besides, wider application of QDs with larger area and high performance like high PLQY requires better manufacture with lower cost and substrates with better flexibility. The idea substrate with flexibility is plastic, but the problem of packaging will also appear. Polymer films and transparent metal films are the possible solutions with both flexibility and transparency. So new synthesis methods should be found to make the mass production of the blue quantum dots LEDs with larger scale[23].

5. Blue perovskite (PVSK) LED

5.1 3D perovskites

A mixed Cl/Br perovskite can have an adjustable luminescence range of 490 nm to 1410 nm by adding more chloride to the material [24]. The first 3D blue perovskite based PeLED has a brightness of 2 cd m − 2 at room temperature, and its EQE is 0.0001%. As can be seen, the pinhole restriction in the film morphology prevents the device efficiency from being optimized. and the gadget may include a shunt path. In addition [7], PCBM is utilized in PeLED devices as an electron transport layer [25].

5.2 2D perovskites

The general formula of 2D perovskite is an aryl or alkyl long chain group with NH32BX4[26]. At room temperature, the researchers found that at 410 nm, the 2D perovskite blue Peled showed a luminescence peak EQE which is 4% [27]. The researchers successfully found the sky-blue particles of ITO/zinc oxide/polyethylene/(4-PBA) 2PbBr4/TFB/MoO3/aluminum by the method of changing the organic interlayer, In this discovery, the brightness of the 2D PeLED is displayed as 186 cd m^{-2}, while its EQE is 0.015%. The researchers expect to realize blue light emission of pure halide perovskite with the formation of quantum confinement effect; However, due to strong exciton phonon coupling, the PLQE of 2D perovskite is limited, which limits the EQE of the blue 2D ring coil[28].

5.3 Colloidal perovskite nanocrystals

Blue CsPb (Br1xClx)3(0x1) PNC was produced by thermal injection, and the resulting blue Peled had an on-voltage of 5.1 V (1 cd m 2), an EQE of 0.07%, and a maximum brightness of 742 cd m 2. They employed poly (9-vinylcarbazole), which has the benefit of avoiding PEDOT:PSS-induced interfacial emission quenching, as the pore transport layer (HTL). PVK, or poly (9-vinyl carbazole), was utilized as a layer to carry holes (HTL). Additionally, this may stop the interface emission quenching brought on by PEDOT:PSS. The NiO2/CsPb(Br1xClx)3 interface was also severely discharge quenched, and NiO2 can be replaced by TFB or PFI (perfluorinated ionomer). With this change, the EQE of dark blue Peled can rise from 0.03% to 0.5%.

6. Conclusion

In this years, blue LED has experienced the development in terms of the luminescent material, luminescence mechanism, and optimization of device structure and material structure. In this paper, focuses on the progress on Blue LEDs, including conventional inorganic GaN blue LEDs, organic blue LEDs, quantum dot blue LEDs and perovskites blue LEDs. The conventional GaN blue LED has been a well-researched and widely used direction with quantum efficiency of 2.7% but the further improvement was limited. The organic blue LED focused on the improvement of the possible

luminescence mechanisms and achieves an internal quantum efficiency of 62.5%. The improvements of the organic approaches will be focused on the enhancing the emission process in each luminance mechanism and avoid the defects and dissociation of the organic layers to extend the lifetime. The traditional Cd based or Pb based quantum dot blue LEDs are toxic. The recently discovered heavy-metal-free QDs can have the EQE of 20.2% and PLQY of 93%, but they still need better manufacture methods to make uniform nanocrystals without too much defects and mature methods for mass production. The carbon based QDs can have the PLQY of 70%±10% and the oleophilic CDs with few oxygens have good development potential. The perovskite blue LEDs have made the EQE higher than 20% but the quantum confinement effect and the emission quenching layer are the mainly limitation that needs more attention on it. In conclusion, the development of blue LED has experienced some great improvement in some new directions and many possible solutions related to the challenges are pointed out.

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