$CsPbX_3$ Perovskite Quantum Dot Laser

Yuhao Wang[1 *]

[1]Department of ECE, University of Michigan, Ann Arbor, US

*Corresponding author: wangyuh@umich.edu

Abstract: Quantum dot laser, which is well known as the 3rd generation of semiconductor laser, has attracted extensive attention of researchers in recent years. Compared with typical semiconductor laser, quantum dot exhibits the characteristics of low threshold, large laser gain, tunable bandgap, which make it promising for laser applications. Among the various quantum dot lasers, perovskite quantum dot laser is one superior type. Perovskite is a group of material with the structure of ABX_3. This group of material is commonly used in solar cell and light emitting device such as perovskite quantum dot blue emitting diode, due to its excellent optical properties of narrow linewidth and high luminance. The perovskite quantum is also found to be a good material of laser gain material. Among all classes of perovskite, $CsPbX_3$ has become an expected material for perovskite quantum dot laser. This work will conclude the theory of quantum dot laser and properties of $CsPbX_3$ quantum dot laser based on current papers and reports.

Keywords: Quantum dot, Semiconductor Laser, Perovskite, $CsPbX_3$.

1. Introduction

Semiconductor was first invented in 1962 and it soon become a significant area in laser field [1]. It is now the most common used lasers in our daily life due to its cheap fabrication process, small size, and reliable structure. In the semiconductor laser, electrons are pumped to the conduction band to achieve population inversion. Then, the process of radiative recombination of electrons and holes will produce photons. One of the key optical properties of semiconductor is the bandgap which determine the wavelength of emitted light. The first semiconductor laser was GaAs based p-n junction laser, which was also a typical semiconductor with the central wavelength of 840nm [1].

In the following years, several approached were made to improve the lasing efficiency, by choosing other materials (e.g., AlGaAs) and optimizing laser structure (e.g., heterostructure, quantum well, quantum dot). The concept of quantum well is evolved from heterostructure, which creates an efficient one-dimension confinement of charged carriers. To provide the confinement, the thickness of sandwiched layer is usually less than 10nm. The concept of quantum dot is raised on the base quantum dot, since it provides a three-dimensional confinement which can be seen as quantum box. The size of quantum of box is extremely small which only contain few atoms or molecules. Thus, in physics, it can be seen as 0D structure. Due to the quantum confinement, carriers are captured in the dot and thus the recombination also happens in the quantum dot. Compared with quantum well laser, the quantum dot laser acts more like gas laser since every single quantum dot can be considered as few atoms or nanocrystal. The advantages of quantum dot laser include [2]: (1) higher temperature operation, (2) less reflection sensitivity, (3) low linewidth enhancement, (4) less defect sensitivity. Due to these advantages, quantum dot laser has become a potential substitute of quantum well laser commercially. However, it also has disadvantages of high nonradiative rated due to Auger processes and weak conductivity of layers when it uses electrical pumping. There are several materials that can be used in quantum dot laser and the most common one is InAs/GaAs. It is dot in well laser, which is embedded in InGaAs quantum well to extend the emission wavelength.

In recent years, perovskite has been considered as a high photoluminescent quantum yield material, which is first used as solar cell. Several research also find it a good candidate of quantum dot laser [2]. It is a group material with ABX_3 structure. This structure is flexible and make it easy to change the size of perovskite nanocrystal. Among all light emitting perovskite, the group $CsPbX_3$ has captured scientists' attention. The $CsPbX_3$ quantum dot has extensive application. At present, $CsPbX_3$ quantum dot can be used as solar cell due to its excellent photovoltaic performance, tunable bandgap

and high Photoluminescence Quantum Yield (PLQY). As light emitting material, LED is also a significant field for $CsPbX_3$ quantum dot. The narrow emission linewidth and other suitable features make $CsPbX_3$ quantum dot the next generation of LED material commercially. As $CsPbX_3$ quantum dot can be used as LED, researchers have thought of the possibility of its application in lasers. As gain material, the $CsPbX_3$ quantum dot has comparably low threshold and higher stability of stimulated emission. For conventional perovskite quantum dot ($MAPbX_3$), the electrical properties of perovskite quantum dot are not stable due to air exposure. Another issue is that due to the accumulation of heat when lasing, the propertied of perovskite quantum dot also change. These issue makes it difficult to operate lasing process in normal air environment at room temperature. To address these problems, the group of $CsPbX_3$ is introduced. It shows the advantages of tunable wavelength, low threshold, and stability in normal air environment.

2. Theory of perovskite quantum dot theory

2.1 Basic laser theory

A laser can be generally considered as a feedback system which includes gain medium and cavity. The gain material will amplify the input signal via the stimulated emission. The cavity will provide optical feedback so that the amplified input signal will oscillate in the gain medium and be amplified through multiple time. In steady state when the intensity of laser in the cavity is stable, gain coefficient is equal to the loss coefficient.

Amplification will happen in the gain medium when it achieves population inversion. In Figure 1, the electrons will be excited from ground state E_1 to excited state E_2 due to external pumping. Thus, the population of electrons in E_2 is large than E_1. When giving a input signal, the excited electrons will decay and generate new photons with same direction and wavelength.

$$E_2 - E_1 = hv \quad (1)$$

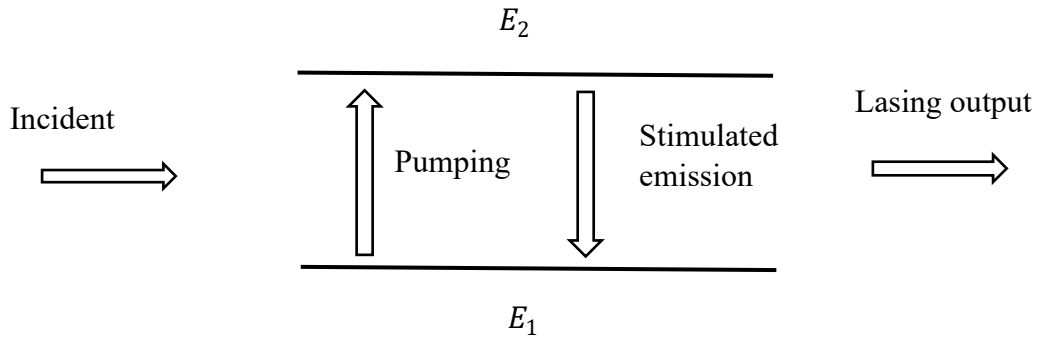


Figure 1. two levels laser diagram

For feedback, there are several types of optical cavity and the traditional one is FB(Fabry-Perot) cavity, which is simple and common. For the design of cavity, two issues should be considered: gain threshold and oscillation wavelength. In the cavity, to generate the lasing output, the start gain coefficient determined by the population difference ΔN should be larger than the loss coefficient. After a period, in steady state, the gain coefficient will finally decrease to loss coefficient which is called threshold gain. For oscillation wavelength, only specific wavelengths are allowed to oscillate in the cavity. For example, in FB cavity, the oscillated wavelengths should satisfy: $\omega_q = q\frac{\pi c}{Ln}$, q is integer; n is refractive index. Thus, the expected output wavelength should be equal or near the oscillated wavelength.

2.2 Quantum dot effect

The concept of quantum dot can be simply considered as a quantum box (cubic) which has quantum size in three dimensions L_x, L_y, L_z. This quantum structure gives electron a 3D confinement.

$$-\frac{h^2}{2m}\nabla^2\psi + V(x,y,z)\psi = E\psi \tag{2}$$

Where,

$$V(x,y,z) = \begin{cases} 0 & 0 < x,y,z < L_{x,y,z} \\ \infty & otherwise \end{cases} \tag{3}$$

The energy of the electron is that

$$E_x = \frac{h^2}{2m}\frac{\pi^2}{{L_x}^2}{n_x}^2, E_y = \frac{h^2}{2m}\frac{\pi^2}{{L_y}^2}{n_y}^2, E_z = \frac{h^2}{2m}\frac{\pi^2}{{L_z}^2}{n_z}^2 \tag{4}$$

The total energy is

$$E_n = E_x + E_y + E_z \tag{5}$$

Similar to the quantum well, the energy level of quantum dot is discrete not continuous. The density of states of quantum dot can be assumed to be delta function

$$\rho = \sum \delta(E - E_n) \tag{6}$$

For the material of quantum dot, it has an energy bandgap E_g when there are massive atoms forming a continuous conduction band and valence band. However, when it is made in the form of quantum dot, there are few atoms and thus the energy level is discrete. The bandgap $E_g{}'$ is

$$E_g' = E_g + E_n \tag{7}$$

As previous description, E_n is determined by the size of quantum dot. When fabricating quantum dot, the bandgap can be easily set to the expected wavelength (usually 1.3 μ m or 1.5 μm, which are suitable for fiber communication) that cannot be achieved by normal semiconductor material, by changing the size of quantum dot. This is significant reason that quantum well and quantum dot laser are commonly used. For all quantum dots, when the size of quantum dot or nanocrystal is less than a specific length which is called exciton Bohr radius. Below this radius, the quantum dot will display obvious quantum confinement. For different material of quantum dots, they have different exciton Bohr radius. For example, for $CsPbCl_3$, $CsPbBr_3$, $CsPbI_3$, their exciton Bohr radius are 2.5 nm, 3.5 nm, 6 nm.

2.3 Quantum dot laser output model

Considering a quantum dot laser with the structure of heterojunction. The P doped material and N doped material serve as cladding layer in the heterojunction. The cladding layers are on the top and bottom of active layer. There are several quantum dots in the active layer which are separated by wetting material. When giving an external pumping (current), the cladding layers will transport the charges into the active layer. The quantum dots will capture those charge in their conduction band and valence band. By the effect of stimulated emission, those charges will recombine in the quantum dot and emit light with the wavelength determined by the bandgap.

The dynamic mechanism of quantum dot is complicated, since it is by various factors including laser structure, broadening, layer density. According to Kathy Lüdge and Eckehard Schöll [3], they gave a dynamic model of a two-level QD injection laser. The theoretical input-output model in steady state is

$$n_{ph} = \eta\frac{\Gamma}{\tau e}\frac{N_{QD}}{N_{sum}}(j - j_{th}) \tag{8}$$

Where n_{ph} is photon density; η is the efficiency of pumping (current injection); τ is total cavity loss; e is the elementary charge; Γ is optical confinement factor which can be simply considered as the ratio of total quantum dots volume divided by active layer volume; N_{QD} is the density of quantum dots which are participated in lasing; N_{sum} is the total density of quantum dots. j is the pumping current density and j_{th} is the threshold current density which mainly depends on the density of charge carrier.

2.4 Perovskite

Perovskite was originally referred to $CaTiO_3$ and was then extended as the term of material with ABX_3 structure, as it is shown in Figure 2. Here, the atom A is monovalent cation including the organic cation methylammonium $MA^+(CH_3NH_3^+)$, formamidinium $FA^+(NH_2CH{=}NH_2^+)$, and metal cation cesium Cs^+. The atom B is divalent metal cation including Pb^{2+}, Sn^{2+}, and the atom X is the halide anion such as F^-, Cl^-, Br^-, I^-.

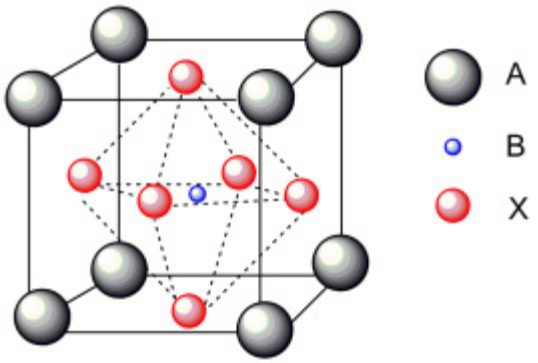


Figure 2. Perovskite Structure [4].

Goldschmidt tolerance factor t and the octahedral coefficient μ are used to measure the structure of perovskite.

$$t = \frac{r_A + r_x}{\sqrt{2(r_A + r_x)}} \tag{9}$$

$$\mu = \frac{r_B}{r_x} \tag{10}$$

r_A, r_B, r_x are the radius ion A, B, X. For perovskite, t should be in the range from 0.8 to 1.0 and μ ranges from 0.41 to 0.89.

When the size of perovskite nanocrystal can be considered as quantum dot, it shows an excellent electrical performance. The photoluminescent quantum yield efficiency can be higher than 90%, which make perovskite a new type of light source material. Perovskite has flexible structure, which means that the cation and anion can be replaced or partly replaced to modify its electrical properties (bandgap, linewidth, absorption efficiency). In the formulas of ABX_3, A cation mainly determined the structure of nanocrystal and electrical properties of perovskite. The radius of A cation determines the lattice of perovskite and the bandgap. And these properties can be changed by replacing A cation with other monovalent cation (MA^+, FA^+, Cs^+) [5]. For other components in perovskite nanocrystal such B cation, it can also be replaced to tune the bandgap and emission wavelength to some extent. The lead ion (Pb^{2+}) is the most common B cation of light emitting perovskite, since it has good stability and PLQY.

3. $CsPbX_3$ Perovskite quantum dot laser

3.1 $CsPbX_3$ as quantum dot

Normal hybrid organic-inorganic perovskite ($MAPbX_3$) is sensitive to moisture and becomes unstable during lasing operation. In recent years, a novel class of perovskite material, the all-inorganic cesium lead halide perovskite ($CsPbX_3$), has been raised into focus. It is more likely to be used in commercial area due to its features of low-threshold and stability, compared with other quantum dot lasers.

$CsPbX_3$ can be used as light source with a very good photoluminescence (PL) property [6]. It has very high PLQY value of 90%, which is larger than traditional CdSe quantum dot and other perovskite quantum dot. Several research have investigated the reason. Firstly, coming back to traditional quantum dot (CdSe quantum dot), it suffers from the size-distribution problem, which causes spectral broadening and generates more trap states. However, in Abhishek Swarnkar [6], $CsPbBr_3$ is not affected by those problems. The PLQY value of $CsPbBr_3$ quantum dot with the size of 11nm is about 90%, which is much higher than CdSe quantum dot, with a narrow FWHM of 86meV. When increasing the temperature, it also shows a good stability of the optical bandgap when the temperature, compared with the red shift of CdSe quantum. Moreover, in the experiment [6], the photoluminescence of $CsPbBr_3$ quantum dot remains stable, without the problem of self-absorption and Förster resonance energy transfer (FERT) [7] which lead to the reduce of PL value. For traditional CdSe quantum dot, it requires a lager quantum confinement to achieve high PLQY with the inhomogeneity of bandgap. However, for $CsPbBr_3$ quantum dot, the experiment shows that, with the size of 11nm, it is enough to achieve relatively high PLQY as well as ensure the homogeneity of bandgap.

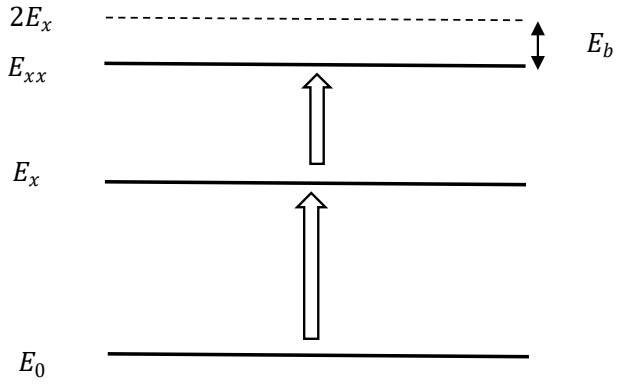


Figure 3. Biexciton mechanism. E_0 is ground state; E_x is exciton state; E_{xx} is biexciton state. The binding energy is $E_b = 2E_x - E_{xx}$

Another reason of high PL is the high binding energy, which makes it easy for the radiative combination of exciton(biexciton) electron-hole pair. Wang et al [8] investigated the $CsPbBr_3$ with the nanocrystal size of 9nm in edge, with central emission wavelength of about 510nm. At room temperature, in air environment, the working mechanism of $CsPbBr_3$ quantum dot's stimulated emission is the combination of excitons and biexcitons. The biexciton is quantum combination of two exciton level (Figure 3) and can be generally expressed in four-level system. The bind-energy of $CsPbBr_3$ quantum dot is about 50meV, which makes $CsPbBr_3$ quantum dot a good material with high PLQY, fast population inversion and high absorption cross-section.

Compared with other perovskite quantum dot, $CsPbBr_3$ quantum dot has relatively higher stability. It shows an excellent stability of stimulated emission (stimulated emission intensity, threshold) in the air exposure environment, compared with the hybrid organic-inorganic perovskite quantum dot such as $MAPbX_3$. In the experiment [8], the duration time of $CsPbBr_3$ quantum dot can exceed 3 weeks.

As a lasing material, the gain and the absorption coefficient of $CsPbBr_3$ quantum dot should also be taken in consideration. Experimentally, it has very high optical gain in green and blue range in

spectral domain, with a cross section of about 2.5×10^{-14} cm^2 at green range, which is higher than that of CdSe quantum dot [8]. For the predominant factor that affects the gain and lasing process of is Auger recombination that generates extra heat rather than photons. The Auger recombination will reduce the lasing intensity and cause heat in the device. The PLQY value is related to such recombination process. For example, with high PLQY value, the material is less affected by nonradiative recombination. For $CsPbBr_3$ quantum dot, the Auger lifetime (105 ps) is almost twice of CdSe quantum dot. However, the gain depletion time is 5 ps which is far below 105 ps, and thus the Auger recombination is less effective in $CsPbBr_3$ quantum dot [8].

3.2 Threshold

The threshold of Perovskite quantum dot laser is determined by gain of $CsPbX_3$ and cavity (DFB, DBR, VCSEL). To generate lasing output, the gain factor g should be larger than the loss coefficient α. The gain factor of $CsPbX_3$ is determined by the level transition and population inversion of $CsPbX_3$. Generally, to achieve high gain, there should be relatively larger pumping power to build large population inversion. The $CsPbX_3$ has a high gain with good efficiency of transferring pumping power to population inversion. To generate output, the gain factor g should be larger than the loss coefficient α which is determined by the optical cavity. When the gain factor is equal to loss coefficient α, this gain factor is called threshold gain and the related pumping power is called threshold pumping. Wang [9] investigated the threshold of the thin film of close-packed $CsPbBr_3$ nanocrystal. With an 800nm 100fs laser pumping source at 1000Hz repetition rate, they found that the output spectral was dominated by spontaneous emission when the pumping intensity was less than 2.5 $mJcm^{-2}$. After increasing the pumping intensity, the output spectral produced a peak at wavelength of about 530nm. This result showed a 2.5 $mJcm^{-2}$ pumping threshold of a $CsPbX_3$ thin film pumping. The performance of $CsPbX_3$ nanocrystal in VCSEL (Vertical cavity surface emitting laser) was also reported [10]. With two DBR layers of 30% reflection on the top and bottom of $CsPbX_3$ nanocrystal film, the result indicated a low pumping threshold of 9 μJcm^{-2}, with a 100fs 400nm laser. This threshold was very low compared with other traditional quantum dots. However, a smaller threshold was reported with the value of 0.39 μJcm^{-2} [11]. They also used DBR VCSEL optical cavity with a 50fs 400nm laser as a pumping source. As discussed above, $CsPbX_3$ has good electrical stability as the lasing threshold linearly changed with the increase of the temperature. From Wang Y et al [12], for $CsPbBr_3$ nanocrystals embedded in Cs_4PbBr_6, in the cavity of two DBRs resonator, the linear range of temperature was 290K~430K. The threshold of 290K was 2.3 $mJcm^{-2}$ while the threshold of 430K was about 2.9 $mJcm^{-2}$. Huang et al [11] also found out a linear range of temperature, from 120K to 300K, with the lowest threshold of 0.15 μJcm^{-2} and the highest threshold of 0.39 μJcm^{-2}. For the whispering gallery microcavity (WGM) laser, $CsPbX_3$ quantum dot is also a good candidate due to its direct bandgap, narrow linewidth. In Yan et al [13], using the pumping source of an 800nm femtosecond laser, the designed a WGM laser based on DA ligand modified $CsPbX_3$ quantum dot with the low threshold a 5.47 μJcm^{-2} compared with 126.81 μJcm^{-2} threshold of unmodified $CsPbX_3$ quantum dot whispering gallery mode laser. In addition, other cavity types are also suitable for $CsPbX_3$ quantum dot. The plasmonic gap-mode cavity reduced the threshold of $CsPbX_3$ quantum dot by coupling the gain medium and cavity [14]. In this work, the $CsPbX_3$ quantum dot was sandwiched between silver nano cube and gold substrate to produce a single $CsPbX_3$ quantum dot laser. With a continuous wave (CW) pumping, the threshold of the laser was about 1.9 Wcm^{-2} (Table 1).

Table 1. Threshold of different type of cavity

Cavity type	Pump source	Threshold
Film [9]	800nm, 100fs	2.5 $mJcm^{-2}$
DBR [10]	400nm,100fs	9 μJcm^{-2}
DBR [11]	400nm, 50fs	0.39 μJcm^{-2}
WGM [13]	800nm, ~fs	5.47 μJcm^{-2}
Plasmonic Gap-mode [14]	CW	1.9 Wcm^{-2}

3.3 Stability

The stability of perovskite of quantum dot during the operation is a significant issue. From previous research, the $CsPbX_3$ quantum dot shows higher stability than conventional hybrid perovskite quantum dot. However, compared with other conventional (gas, semiconductor) lasers, the stability of $CsPbX_3$ quantum dot is still poor, due to the thermal condition and moisture environment. Several approaches are made to optimize the stability of $CsPbX_3$ quantum dot. The stability of $CsPbX_3$ quantum dot is thermal dependent since the thermal energy will affect the process of Auger recombination in $CsPbX_3$ quantum dot and will be possible to cause the decomposition of quantum dot. According to Huang et al [11], the experiment used the fs scale optical pumping and ns scale optical pumping to test the stability of $CsPbX_3$ quantum dot. The result showed that with the fs scale pumping, the output intensity was more stable. Therefore, when designing $CsPbX_3$ quantum dot laser, the thermal energy attained from pumping should be considered.

Some methods such as passivation are introduced to improve the stability of $CsPbX_3$ quantum dot laser. One passivation method [15] is that the surface of $CsPbX_3$ quantum dot is wrapped by an organic-inorganic pair (DDAB). This approach improves the PLQY and stability obviously. There is a big loop of emission intensity peak. After 4-month exposure, the $CsPbX_3$ quantum dot's PL intensity curve versus pumping remain its tendency and shows the nearly same threshold.

The glass embedded $CsPbX_3$ quantum dot also exhibits high stability [16]. This method efficiently protects the quantum dot from the external environment. Apart from improving stability, it also offers function of water resistance Although the hold quantum dot is embedded in glass matrix, its photoluminescence properties remain. The process of making glass embedded $CsPbX_3$ quantum dot utilize melt-quenching process. First, the glass and $CsPbX_3$ quantum dot's chemical components are mixed. Then, with the increasing temperature, the glass starts to melt. During melting, the chemical components of ion will diffuse into glass and be finally uniformly distributed in the glass. After quenching, these components ions are fixed. Through the following heating and crystallization process, the $CsPbX_3$ quantum dots are finally formed in the glass. Comparing with $CsPbX_3$ quantum dot without glass embedded, the type of glass relatively affects the properties of quantum dot. The glass matrix has significant impact on the morphology of quantum dot, which changes the quantum confinement and wavelength of quantum dot. The optical loss is determined by the difference of refractive index between quantum dot and glass. To avoid large scattering effect, the refractive index of quantum dot should be close to that of glass.

4. Conclusion

Quantum dot laser is expected to be the next dominant type of laser in commercial application due to its tunable bandgap and low threshold. This work generally describes the theory of quantum dot laser and illustrates one potential class of perovskite ($CsPbX_3$) material as quantum dot. After this study, several reasons why choose $CsPbX_3$ other than other conventional quantum dot material were concluded. Firstly, comparing with convention semiconductor quantum dot, it has high PLQY and narrow line width. Comparing with other perovskite material, it is more stable. The $CsPbX_3$ quantum dot laser can have various types of optical cavity. This work briefly lists the threshold behavior of different cavity (VCSEL, DFB, DBR, WGM). The stability of $CsPbX_3$ quantum dot cannot be neglected. Several approaches are introduced including passivation and glass embedded methos,

which improves the photoluminescence properties and stability of $CsPbX_3$ quantum dot. After the discovery of $CsPbX_3$ quantum dot, it has captured increasingly attention in recent ten year. It is potential to replace the traditional semiconductor in devices such as cellphones. The problem of stability still limits its applications and further researches are still needed to realize the commercial application in the future.

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