Light-Emitting Devices – Luminescence from Low-Dimensional Nanostructures

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1. Introduction

Luminescence is the emission of light that does not cause energy loss from the temperature of the emitting material. The emission of light occurs because of an excitation in the material and has different forms like photoluminescence, electroluminescence, thermoluminescence, etc. Reasonable efficiency, high stability, and easy and economical fabrication methods make light-emitting devices a good choice for mass production.

This chapter focuses on the luminescent properties of low-dimensional nanostructures and reviews the principles of luminescence. Different materials for this application and some of the best-known electroluminescent devices are reviewed. In addition, low-dimensional nanostructures, a simple method for preparing them, and the development of these structures for application in light-emitting devices are briefly described.

2. Fundamentals of luminescence

In this part of the chapter, the basis of radiation from solids that produce visible light is discussed. Luminescence is the emission of light by an excited substance. In order to create an emission, an electron needs to be excited from the ground state (E_1). During transition of the electron from the excited (E_2) to the ground state, a photon is released. In order to start this transition, we need to stimulate the electron in the excited state. This process is shown in Figure 1:



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Figure 1. Transition of an electron from excited (E2) to ground (E1) state in a double-state system that results in the release of a photon.

In semiconductors, the ground state is usually referred to as electrons in the valance band while excited state electrons are known as the conduction band. Unlike the metals in semiconductors, these two states are separated by an energy gap called the bandgap (E_g). Therefore, a minimum energy of the bandgap is necessary to excite an electron from the ground to the excited state. Luminescence from semiconductors can be observed by exciting the electrons to higher states like the conduction band and subsequent decay to the ground state. There are different methods of providing the excitation that cause luminescence from a material. Depending on the excitation origin, there are several types of luminescence such as photoluminescence, electroluminescence, cathodoluminescence, chemiluminescence, thermoluminescence, etc. When an electron is promoted from the valance band to the conduction band, a hole will remain in the valance band [1].

If a semiconductor absorbs the electromagnetic radiation of a photon, an electron in the valance band can be excited to the conduction band. When the excited electron returns to a lower state, it causes the radiation of a photon in a process called photoluminescence (PL). When electrical potential is applied, the conversion of energy from electrical energy into light emission is called electroluminescence and the device that produces it is called a light-emitting diode (LED). Due to the narrow nature of the bandgap in elemental semiconductors, they are not suitable candidates for LEDs. Instead, other semiconductors with wide bandgaps are frequently used for this application. The term 'wide bandgap semiconductors' describes those that exhibit light in the visible part of the light spectrum and in the shorter wavelengths of ultraviolet.

Compound semiconductors from group III-V (such as III-nitrides), group II-VI (such as oxides and chalcogenides) are the most important types of wide bandgap semiconductors, although other semiconductors like SiC, Si, and Ge are also studied for light-emitting devices. Figure 2 shows some of the semiconductors that have been used for light-emitting devices. The materials with small lattice constants have stronger interatomic forces and their outer electrons are strongly bound to the lattice, which means that such materials have a large bandgap energy. By changing the structure and doping, many colors can be provided for LED applications. While the infra-red and red LEDs have been studied for 40 years, bright blue, violet, and UV LEDs have been the subjects of research studies in more recent years.

Even more recently, light-emitting devices based on polymer materials have been investigated. In the next section, polymer and organic light-emitting devices are discussed briefly.



Figure 2. A selection of semiconductors for light-emitting devices. The area between 2 and 3eV are distinguished for visible wavelengths.

3. Light-emitting devices

In this section, different types of light-emitting devices and their working principles are summarized; they are grouped based on the type of material that is applied to create the light emission.

3.1. Emission from inorganic semiconductors

To provide LEDs with different colors, III-V and II-VI materials like GaAlAs, AlinGaP, InGaN, AlN, ZnO, and ZnSe are typically used. The p-n junction is situated between two electrodes, at least one of which should be transparent. Industrial LED applications are highly focused on white color as a light source and as backlighting for electronic devices. New types of LEDs with white color have been developed by mixing luminescent materials in three red-greenblue (RGB) colors. There are two common methods for the generation of white color in LED devices.

The first method uses an individual combination of red, blue, and green LEDs to mix the provided colors and produce the white color shown in Figure 3a, while the second is a combination of different phosphorous materials to generate the white color shown in Figure 3b.



Figure 3. Mechanism of providing light-emitting diode with white color (a) Combination of three red-green-blue (RGB) LEDs (b) Mixing of different luminescent materials to generate the white color.

3.2. Emission from small organic molecules

The first emitting organic material was observed by Pope et. al [2] and led researchers to investigate organic materials as light-emitting devices [3-6]. The working principles of these devices are similar to normal LEDs with the difference that an organic compound is used as an emissive layer. Such devices are called OLEDs (organic light-emitting diodes). A typical OLED consists of at least one emissive layer and one conductive layer which are deposited between two electrodes (anode and cathode). Like semiconductors, these organic materials show other conductivity characteristics under an electrical field between two electrodes. Analogous to p-n junction semiconductors, the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) are regarded as the valance and conduction bands, respectively, of inorganic semiconductors [7].

Figure 4 shows the basic schematic structure of an OLED, which can also consist of an electron transport layer and hole transport layer. At the first stages of the fabrication of OLEDs by Philips, the devices consisted of simple structures of poly(dialkoxy-p-phenylenvinylene) (dialkoxy–PPV) that were situated between a metallic electrode and tin-doped indium oxide (ITO) as the transparent conductive layer [8]. Glass is extensively used as a substrate to fabricate this type of LED, while polyethylene terephthalate (PET) can also be used for fabrication of flexible devices [9, 10].

There has been extensive effort to fabricate OLEDs with primary colors that involved studying different organic materials. H. Fukagawa et. al. [11] reported highly efficient red phosphores-



Figure 4. The basic schematic structure of an OLED consist of electrodes (cathode and anode), organic luminescence layer, and transparent conductive material.

cent OLEDs fabricated using platinum complexes as dopants, but most of the studies were performed using iridium complexes and proposed novel complexes of platinum and a useful host/dopant combination. Many host/dopant combinations have been investigated in an effort to provide red OLEDs that show high stability. The optimized device exhibited good color saturation and high efficiency as well as working with low driving voltage. The lifetime of the fabricated devices was estimated to be approximately 10,000 hours.

Green OLEDs with high efficiency, using copper complexes, were reported by S. Igawa et. al. [12]. The copper complexes are an inexpensive emissive choice and exhibit substantial thermal stability compared to other rare earth metals. They exhibit strong green emission at room temperature with a wavelength in the range of 523-544 nm. A conventional OLED structure containing electron and hole transform layers, polymer complex, and electrodes shows the bright green emission with quantum efficiencies between 11.9% and 17.7%.

The complexes that can provide blue colors are more attractive than the red and green complexes for application in OLEDs because of their short wavelength emission. Several organic emissive materials have been used in blue OLEDs. A novel benzofuropyridine complex was synthesized by Chil Won Lee et. al. [13]. These OLEDs were fabricated with two different methods of spin-coating and a vacuum-based route. Both devices exhibited an emission peak at 486 nm, with a strong shoulder peak at 493 nm also observed due to the thickness of prepared thin films. They achieved high quantum efficiencies between 18.0% and 25.0%, but they did not report the lifetime of their fabricated devices.

By mixing these three main colors, white color can be created with OLEDs. The pioneer research study for these LEDs was initiated by Kido et al. [14] by mixing three dyes (blue, green and orange) into the emissive layer to obtain the white color.

3.3. Emission from large polymer molecules

In addition to small organic molecules, polymers can also be used as an electroluminescent material. Due to the variety of polymer molecules, there is great variability in the emission wavelength of polymer light-emitting diodes (PLEDs) [15-17]. Most of the conjugated polymers are soluble in water-based solutions and therefore can be deposited with wet chemical coating methods like dip-coating, spin-coating, and printing, which makes them highly suitable for the fabrication of devices on big substrates [18]. Additionally, by using flexible substrates, flexible devices like displays can be created. However, because of the degradation of the polymers by oxygen, such devices should be encapsulated and therefore have shorter lifetimes compared to inorganic LEDs.

A lot of research has been done to increase the stability of PLEDs [19-20]. A. Berntsen et al. [19] improved the stability of polymer LEDs operating at daylight under ambient conditions. The lifetime about 5000 hours is obtained and under higher temperatures (70°C), they have one tenth of this time. Y. Cao et al. [21] applied an ultra-thin alkaline earth metal as electrodes for PLEDs and increased their stability and invented this method to increase operating life of PLEDs in which the decay of efficiency and light output is substantially postponed.

Due to the diversity of organic materials, PLEDs can also create different colors from blue to red in visible wavelengths. Several polymer emitters can provide the three primary colors. The common approaches to fabricate the white light OLEDs include doping with small fluorescent or phosphorescent combinations, polymer blending, fabrication of devices with multiple emissive layers, and synthesize a single polymer that can provide the white color [22]. In [22], the authors reported new design techniques for high performance white PLEDs. So far the external quantum efficiency more than 20% has been obtained for these devices and the total power efficiency more than 80 lm/W has been achieved. By improving and increasing the lifetime of these structures, they can be used as energy-saving light sources (for general light, because of low energy consumption), back light for electronic devices (because of low thickness), and flat or flexible displays.

3.4. Emission from phosphor particles

Theoretically, the electroluminescence (EL) from phosphor materials is classified into two groups: injection EL and high field EL. The high-field EL can be further grouped into different types in terms of the phosphors (powders vs. thin film) and the voltage (DC vs. AC). Due to the requirements for various applications, injection EL (high intensity lighting-emitting diodes (LEDs), for example,) have a wide application in liquid crystal displays (LCDs) backlighting and are entering the lighting market.

Luminescence is the emission of lights that does not derive energy from the heating of emitting material. Reasonable efficiency, high stability, and an easy and economical fabrication method make these devices a good choice for mass production. Electroluminescent lamps can be made by sandwiching a phosphor material between two conductive electrodes. At least one of these electrodes should be transparent (TCOs like ITO, FTO or AZO layer). An EL lamp consists of phosphor material; Dielectric layer (e.g. barium titanate with a high dielectric constant to

increase the electrical field between the electrodes); Conductive layers (e.g. ITO as transparent layer and silver coating as back contact); Barrier layer to keep phosphor from moisture, and dust. A schematic structure on an EL lamp is shown in Figure (5).



Figure 5. Structure of EL lamp.

By applying a high electrical field (provided with an AC voltage through the conductive electrodes) over the phosphor particles, the electrons will be trapped in the interface layer and will be tunneled to the conduction band of the phosphor layer. The accelerated electrons by electrical field (with enough kinetic energies) excite the impurities from the ground state to excited state. When the electrons come back to their ground state, light is emitted. For this purpose, the electrons must be able to localize in the ground state; otherwise, having a high electric field, they do not emit light. The phosphor material for application in EL devices should be an insulator and contain impurities. It also has to be transparent for the emitted wavelength.

Zinc sulfide is one the most important candidates for phosphor material that have been doped with different chemical elements like magnesium, manganese, copper, or vanadium to provide a different range of colors. Due to the simple structure, they can be used for applications in back-lighting but are still not useful for general light applications.

4. Low dimensional nanostructures

Many physical and chemical properties of materials can be changed effectively by controlling the size of materials between 1 and 100 nm. In such nanostructures, the finite size can generate novel optical, electrical, or magnetic properties that cannot be observed in bulk materials. In other words, the atoms of material on the surface show different properties and therefore by decreasing the dimensions of a material, the high surface-to-volume ratio enables them to exhibit new properties.

Nowadays, semiconducting nanostructures have been the subject of great interest due to their unique optical properties. With more investigation and development into such structures,

many optoelectronics devices have been produced. Materials with a large surface area-tovolume ratio can be a good candidate in optoelectronics applications. Applications of devices based on optical properties can include lasers, solar cells, optical detectors and sensors, displays, photo-catalysis, imaging and phosphor devices [23-30].

Various methods have been used for the fabrication of nanostructures, like molecular beam epitaxy (MBE) [31,32], sol-gel [33,34], chemical vapor deposition (CVD) [35,36], lithography [37,38], physical vapor deposition (PVD) [39], and chemical methods [40-41]. Among these methods, the low cost and high efficiency technologies have attracted more attention.

In this section, we present a low-cost and industrial method to obtain such nanostructures with a chemical vapor deposition method by an electrical furnace. In addition, their structural and optical properties are studied. So far, a large number of methods have been used to synthesize wide bandgap semiconductors. Some of the currently used methods have problems regarding high temperature, high vacuum conditions, the necessity of using expensive equipment, and difficulties with quality or commercialization. The focus of this chapter is on development of a low-cost method for producing high quality nanostructures with regard to facilities and experimental conditions.

This method is based on the evaporation of source materials in the hot-zone of a horizontal electrical furnace, reacting with an active gas and finally transporting on the substrates in cold-zone of the furnace using a carrier gas. Despite the simplicity of this method, there are a lot of parameters that can affect a growth mechanism. Among these parameters, the effects of growth conditions such as synthesis temperature, temperature rate, amount of carrier and active gases, source chemical materials, and impurities on the structural properties of these nanostructures are studied.

The variation of these parameters leads us to the systematic modification of one and two dimensional (1D & 2D) nanostructures such as nanowires, nanorods, nanosaws, nanodendrites, nanobelts, and nanosheets. Reasonable emissions of these structures in the blue-green, violet, and ultra-violet (UV) wavelengths can lead us to produce light emitting devices. Also the effect of structural properties and impurities are investigated.

A horizontal furnace is used in this experiment, which is divided into two independent zones: zones (a) and (b), called the hot and cold zones, respectively. Active and carrier gases are introduced through the tube on one side in order to produce the active gas system and carry them to the cold zone of the furnace. The other side of the chamber is evacuated using a vacuum pump. Source materials with the desired molar ratio are inserted into the center of the hot zone. The substrates can be placed at different distances from the source material in the cold zone. Before increasing the temperature, the chamber is flashed with inactive gases (like argon) to remove the residual oxygen from the air. The temperature of the hot zone is increased to, and kept at, different temperatures for various durations. Finally, a thin layer of synthesized material is deposited on the substrates in the cold zone. The distance between the source material and the substrates and also the temperature of the substrates are two important parameters that should be controlled during the deposition. The schematic setup of this furnace with a quartz tube is shown in Figure 6.



Low dimensional materials attract more interest these days for applications as opto-electronic devices. They have a high surface-to-volume ratio compared to bulk materials and it causes them to show unique properties. When we talk about the low dimensional materials, they can be categorized as: two-dimensional (2-D) like thin films and quantum wells, one-dimensional (1-D) like nanowires, nanorods or quantum wires, and zero dimensional nanostructures (0-D) like quantum dots. Each category shows interesting optical properties [42]. The emission behavior of such nanomaterials is much more complicated than the bulk materials.

The optical properties of nanomaterials depend on many parameters like dimensions, size, shape, temperature, and other variables like dopants and impurities. Even surface characteristics and the surrounding atmosphere have a strong effect on optical properties of these materials. So by controlling these properties, one can generate optical devices covering the whole visible and UV regions of the optical spectrum. The red-shift or blue-shift of photoluminescence spectra of semiconductor nanoparticles is one the most famous examples of changes in optical properties of the materials. In semiconductors, size is the most important parameter to affect the optical properties.

A. Wolcott et. al [43] studied the optical properties of CdTe quantum dots and the results showed different emission colors from CdTe quantum dots. The emission of different colors changes for the particles with different sizes. Increasing the size of the nanoparticles (that were obtained by changing the refluxing time) causes the emitted light to change from blue to red. Also A.M. Schwartzberg showed different colors emitted by hollow gold nanospheres. By varying the diameters and wall thickness, the samples produce different colors from red to violet [44].

Two kinds of wide bandgap semiconductors, such as zinc oxide (ZnO) and aluminum nitride (AlN), are the main subject of this chapter. These nanostructured wide bandgap semiconductors show good light emitting properties in blue-green, violet and ultraviolet (UV) wavelengths. As we discussed in the last section, by controlling the growth condition in this method, we can have different nanostructures with various sizes, dimensions, defects, shapes, dopants

and crystallinities. Changes of the surface-to-volume ratio with size and quantum confinement effects cause dependency of the optical properties on the size and dimension.

ZnO is a wide bandgap semiconductor with a direct bandgap of about 3.2 eV at room temperature. Because of the unique optical properties, this material has been used as a suitable semiconductor for different optical devices like UV light emitters, lasers, and detectors. Because of the wide range of synthesis methods, ZnO can be grown in different shapes and structures like one or two-dimensional nanostructures. Structure and doping are two important parameters that have a strong effect on optical properties of this material. One of the major aspects of growth of ZnO is doping of it with different metals to approach an n-type semiconductor.



Figure 7. ZnO nanostructures with different shapes to study the effect of shape and size on luminescenct properties (Reprinted from [45] with permission from Elsevier).

In the previous work [45] by changing the growth condition, we prepared different ZnO nanostructures. Nanowires with diameters of 20 ± 5 nm are observed in Figure 7(a). In addition, as the amount of oxygen in the carrier gas mixture increases, the nanostructures change from wire structures to nanorods as shown in Figure 7(b). Figures 7(c–e) show the 2D ZnO nanobelts (with a typical width in the range of 100 to 200 nm), nanodendrites, and nanosheets (typically 10–20 nm in thickness and 1–2 µm in width), respectively, grown on the Si substrate.

After the excitation of an electron with an external energy, electrons and holes possess higher energies and they will recombine together and form an exciton which has a lower energy state. The released energy from this recombination can result in the emission of photons or Auger electrons [7]. In photoluminescence, the recombination of electron and hole cause the radiation of a photon. In semiconductors, these radiations may result from the near band emission transitions and from the defects in the structures.

To explain this fact, we will return to the PL emission from the nanostructures in Figure 7. Figure (8) shows the photoluminescence spectra of the crystalline structure at the related spots. Three main peaks are observed. In addition to an emission in the UV region (λ =380 nm), broad visible emission is observed with two main peaks at ~485 and ~530 nm. The origin of UV emission is radiation because of the recombination of electrons near or in the conduction band with a hole near or in the valance band, which is called near band emission. Due to the bandgap of ZnO (3.37 eV at room temperature), this emission is in the ranges of UV wavelengths. The visible emission from the ZnO nanostructures has been investigated in many studies [46] and this green luminescence in ZnO was attributed to oxygen vacancy, zinc vacancies, interstitial oxygen and zinc atoms, substitution of oxygen at zinc atom positions and donor-acceptor complexes.

The intensity of emissions is dependent on the nanostructure's size. The UV emission was stronger than the visible emission for samples with larger ones. Below a certain size, the luminescent properties of ZnO nanostructures should be dominated by the properties of the surface. An enhanced deep-level emission for thinner nanostructures has been observed and attributed to their larger surface area. As we discussed, the green light emission intensity progressively increases, in proportion to the UV emission, as the nanostructure dimensions decreases. This suggests that there are a large number of oxygen vacancies in the nanowires. Generally, the defects are present at the surface of the ZnO structure. ZnO nanowires with a small diameter have a higher surface area-to-volume ratio, which results in a high level of surface defects. Therefore the visible emission intensity increases.

The effect of doping was also investigated on luminescence properties of ZnO nanostrustures. The ZnO nanostructures grown by this method are doped by only a few percent of copper and iron as the details are discussed in [45]. The photoluminescence from ZnO:Cu can generally be similar to pure ZnO nanostructures underlying physical mechanisms as free and defectbound excitons, deep and shallow donor–acceptor pair recombinations, and deep-defect associated emissions. A low-intensity shoulder peak at about 455 nm has also been observed as shown in Fig. 9(a).



Figure 8. Photoluminescence spectra of ZnO nanostructure with different shapes and sizes (Reprinted from [45] with permission from Elsevier).

The recombination of donor–acceptor pairs involving Zn²⁺and Cu⁺states is known to yield a blue-green emission. This recombination is responsible for the increased defect luminescence in the ZnO nanostructure. The peak centered at 455 nm can be attributed to Cu²⁺–Cu⁺transitions where the hole remains localized on the Cu⁺center [47,48]. In the case of the introduction of iron as a dopant, the UV emission band of ZnO has a red shift (from 385 to 485 nm) as well as the green emission band showing a red shift (from 530 to 542 nm) as shown in Fig. 9(b).



Figure 9. Photoluminescence spectra of ZnO nanostructure with a few percent of (a) copper (b) iron as dopant (Reprinted from [45] with permission from Elsevier).

III-nitrides are also interesting materials for application as light emitting materials. As another wide bandgap semiconductor, AlN, with a bandgap of about 6.1 eV at room temperature, will be introduced. AlN is a direct bandgap semiconductor with a bandgap of about 6.1 eV at room temperature, the largest among semiconductors. Thus, AlN emitters are expected to be good candidates for ultraviolet and blue-violet optoelectronics devices [49]. This work emphasizes novel results on the growth and optical properties of an aluminium nitride (AlN) nanostructure by direct nitridation. The nitridation process was done in a horizontal tube furnace at different temperatures by introducing an N_2 gas flow. Most of the last efforts for synthesizing AlN nanostructures [50, 51] are performed using ammonia gas as this gas is toxic and corrosive.

Among the synthesis methods for AlN nanowires, the direct nitridation of metallic aluminium powder has been attractive because of the low cost of raw materials and the simple nitridation setup. At the beginning of growth, the reaction temperature is higher than the aluminium melting point, so the Al is in liquid form. These liquid droplets cause the aggregation of obtaining nanostructures into spherical islands. However, we have overcome this problem by using ammonium chloride (NH₄Cl). The addition of NH₄Cl to the starting Al powder produced many pores because of the decomposition of NH₄Cl [52-54]. Therefore, the introduction of porosity during the synthesis can enhance the nitridation rate because N₂ gas easily has more access to the pores, which causes the formation of nanostructures AlN powders with low agglomeration. Different AlN nanostructures (Figure (10)) were synthesized and analyzed by scanning electron microscope.



Figure 10. SEM images of AlN nanostructures grown by direct nitridation of aluminium powder. (a, b) AlN nanowires with diameters less than 50 nm (c) AlN nanotips, and (d) long AlN nanowires with diameters about 1 micrometer.

The photoluminescence (PL) spectra were obtained by a 325nm He-Cd laser as the excitation source measured. The results show high intensity light emitting emissions in blue and violetblue wavelengths for these structures at room temperature. As shown in Fig (11), a typical PL spectrum of these nanostructures shows a peak at 450 nm. Photoluminescence properties of these structures are investigated aiming at the potential application in the field of blue and violet-blue light emitting diodes (LEDs) [55].



Figure 11. Photoluminescence spectra of AlN nanostructure which shows a broad peak at 450 nm.

6. Conclusions

This chapter was focused on light emitting materials and devices. The working principles of light emitting devices are discussed and different materials for this application are introduced. Different types of light emitting devices, including inorganic light emitting diodes (LEDs), organic light emitting diodes (OLEDs), polymer light emitting diodes (PLEDs), and electro-luminescence (EL) lamps are compared.

Different types of the inorganic semiconductors from group III-V (such as GaN, AlN), group II-VI (such as ZnO, ZnS) are the most important wide bandgap semiconductors, for lightemitting devices. In addition, several organic and polymer complexes that can provide the main three colors have been used in OLED and PLED deives. Due to the wide applications of white color LEDs, different methods and different studies for fabrication of these devices are discussed. Stability of light emitting devices is one the most concerns of the providers. A lot of research has been done to increase the stability of these structures under ambient conditions. The lifetime about several thousand hours is obtained. Electroluminescent lamps are other applications of light emissive materials. Luminescence is the emission of lights that does not derive energy from the heating of emitting material. Therefore due to their efficiency and high stability, these devices are good choices for mass production.

A simple chemical method based on vapor transport is applied for fabrication of semiconductor nanostructures. Nanostructures with different shapes and structures offer several advantages for light emitting devices and are receiving increasing attention as a light emissive material to improve the efficiency of the optical devices. These structures could be developed to overcome the problems for fabrication of UV and blue-violet light emitting devices.

ZnO is a wide bandgap semiconductor which because of the unique optical properties, has been used as a useful semiconductor for the optical devices like light emitters, lasers, and optical detectors. Because of the wide range of synthesis methods, it can be grown in different shapes and structures from one-dimensional structures like nanowires and nanorods to two dimensional nanostructures like nanosheets and nanodendrites. AlN with a direct bandgap semiconductor of 6.1 eV at room temperature, the largest among semiconductors, is one the most applicable semiconductors for application in light emitting device and the AlN nanostructures with different shapes and sizes are expected to be good candidates for optoelectronics devices. ZnO and AlN nanostructures are synthesized using a chemical vapor condensation method and their luminescent properties are investigated at roon temperature.

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