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Structural Properties, Energy Interaction, and Applications of CdS Nanomaterial Doped with Rare-Earth Ions

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Spectroscopic analysis of trivalent praseodymium (Pr^{+3}) and neodymium (Nd^{+3}) ions have received very much interest and reveals good fluorescence efficiency in visible and near IR (NIR) region. Cadmium sulfide (CdS) nanomaterial sample doped with Pr^{+3} and Nd^{+3} ions are synthesized at room temperature by simple chemical precipitation method. The synthesized sample has been characterized by powder X-ray diffraction studies, scanning electron microscopy, transmission electron microscopy, and energy dispersive X-ray spectra. Absorption spectra of CdS:Nd^{3+} and CdS:Pr^{3+} nanoparticles have been recorded at room temperature. Various parameters such as interplanar spacing, micro strain, dislocation density, distortion parameter, and particle size of nanomaterial are computed. TEM and XRD analysis reveal the spherical structure of the CdS:Nd^{3+} and CdS:Pr^{3+} nanoparticles in the prepared samples. Energy interaction parameters of the CdS:Nd^{3+} and CdS:Pr^{3+} nanoparticles are computed using UV-visible absorption spectrum. Actually, CdS nanoparticle amplifies the luminescence intensity of Pr^{+3}, Nd^{+3} ions by up conversion processes. So energy transfer can be observed from CdS nanoparticles doped with Pr^{+3}, Nd^{+3} ions. Therefore, the use of pump sources and diode lasers in the UV range can be used to excite these samples to work as lasers. CdS:Nd^{3+} and CdS:Pr^{3+} nanoparticles find potential application as a window material for hetero-junction solar cell, light emitting diodes (LED), biological sensors, address decoders, gas detectors, opto-electronic devices, etc. CdS nanomaterials are also used as pigment in paints and in engineered plastic due to their good thermal stability.

1. Introduction

Study of nanoscience and nanotechnology is related to the use of the familiar properties of materials smaller than 100 nm to create new useful objects. The most common working definitions of nanoscience and nanotechnology as given by the Royal Society and Royal Academy of Engineering UK are as the following: "Nanoscience is the study of phenomena and manipulation of materials at atomic, molecular and macromolecular scales, where properties different significantly from those at a larger scale and nanotechnologies characterization production and application of structures devices and systems by controlling shape and size at nanometer scale."[1] This work is made possible by being able to manipulate structures at the size-scale of atoms. Nanotechonolgy is an emerging, interdisciplinary area of research with important commercial applications,[2] and will, most assured by, different dominant technological field in new world economies. Few such application of nanotechnology is schematically illustrated in Figure 1.

This intense interest in the science of the nanomaterials, which confined within the atomic scales, stems from the fact that this nonmaterial exhibit fundamentally interesting unique properties with great potentials of next generation technologies in emerging fields of the modern era like electronics, optoelectronics, computing, biotechnology, medical imaging, medicine drug delivery, structure of materials, aerospace, and other engineering fields and many more.

All these fields are basically utilizing semiconductor nanoparticles of certain specified characteristics derived by mixing a proper fraction of dopant material with it to employ it in a well-defined sub domain of usage. The subsequent Section 2 of the paper will discuss the detailed properties essential for such semiconductor nanoparticles.

2. Semiconductor Nanoparticles

Nanoparticles useful for developing the desired nanosystems of modern world are basically developed from the group II–VI semiconductors. Semiconductor nanoparticles are thus a type of compound semiconductor composed of group II–VI elements of the

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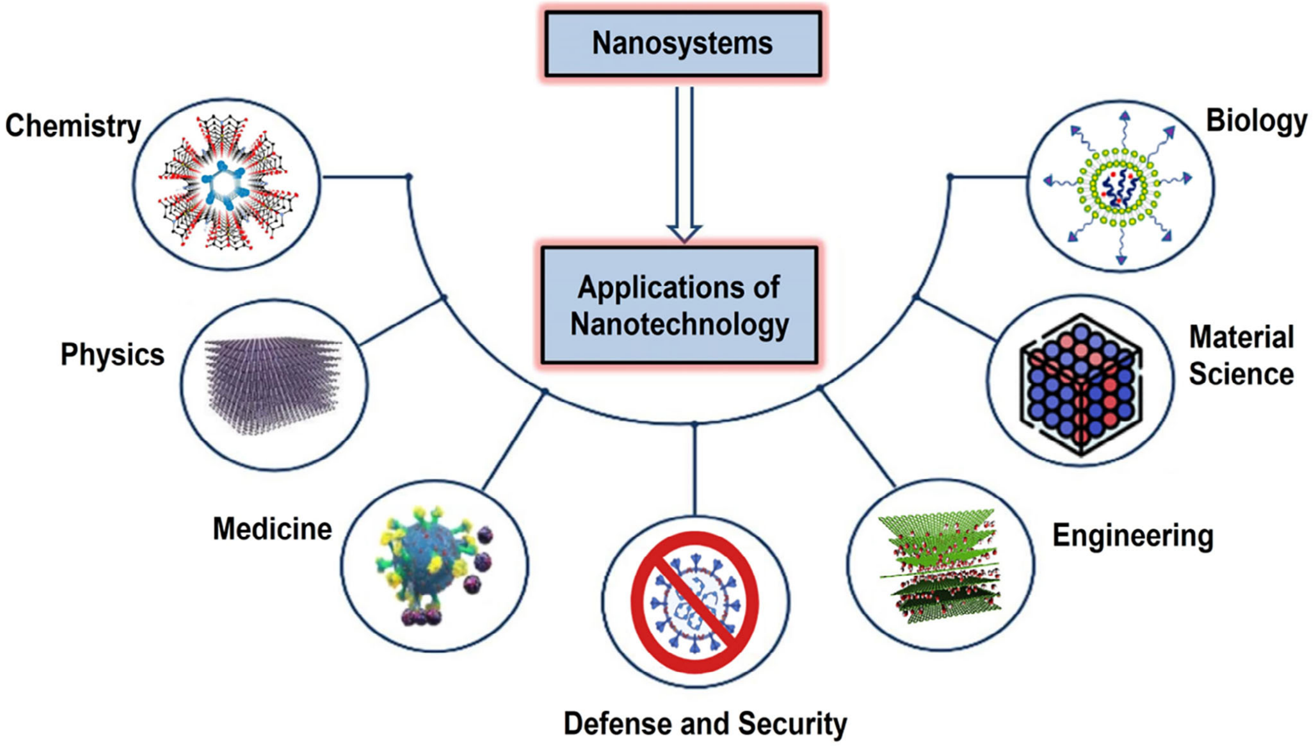

Figure 1. Applications of nanotechnology in different nanosystems used in the fields of chemistry, biology, physics, material science, medicine, engineering, defense, and security.

Table 1. Properties and applications of some semiconductor nanoparticles.

Semiconductor	Band gap	Properties and potential applications
Cadmium sulfide (CdS)	2.42 eV	Direct band gap, used in photoresistors and solar cells, CdS/Cu_2S was the first efficient solar cell. Commonly used as quantum dots when doped material can act as a phosphor.
Cadmium telluride (CdTe)	1.49 eV	Direct band gap, used in solar cell with CdS, used in thin film solar cells and other CdTe photovoltaics less efficient than poly-silicon but is a cheaper fluorescent material which can be used at 790 nm.
Cadmium selenide (CdSe)	1.74 eV	Direct band gap, intrinsic n-type, difficult to dope as p-type, doping with nitrogen is possible, can be used in optoelectronics, tested for high efficiency solar cell.

periodic table. These particles are having a wide and direct band gap structures.

The physical properties of such materials depend on their crystallite sizes and they actually show size dependent electrical and or optical properties in the quantum size region. Due to the fundament as well as technological importance, the modification in the energy band gap of semiconductors is the most attractive property. These are very important in many fields, due to their tunable electrical and optical properties. Semiconductors which possess such property of tunable energy band gap are considered to be the materials for next generation flat panel displays, photovoltaic devices, optoelectronic devices, laser devices, photonic band gap devices, etc. Some of the well-known group II–VI semiconductor materials useful for developing nanosystems are: cadmium sulfide (CdS), cadmium selenide (CdSe), cadmium telluride (CdTe), mercuric sulfide (HgS), mercury selenide (HgSe), mercury telluride (HgTe), zinc oxide (ZnO), zinc selenide (ZnSe), zinc sulfide (ZnS), etc. The properties and potential applications of some of the above semiconductors are given in Table 1.[3,4]

Generally, nanomaterials have structured components with at least one dimension less than 100 nm (1 nm = 10^{-9} m) and having distinctly different physical and chemical properties in comparison to their micron size counterpart.[5] In nanoparticles the various material properties such as electrical, mechanical, optical, magnetic, etc. can be selectively controlled by engineering the size, morphology of the materials; using a variety of synthesis methods, in the various forms like thin films, powder quantum wires, quantum wells, quantum dots, etc. Nanocrystals are characterized as atomic clusters and are called quantum confined systems.[6] The requirement for quantum confinement is that

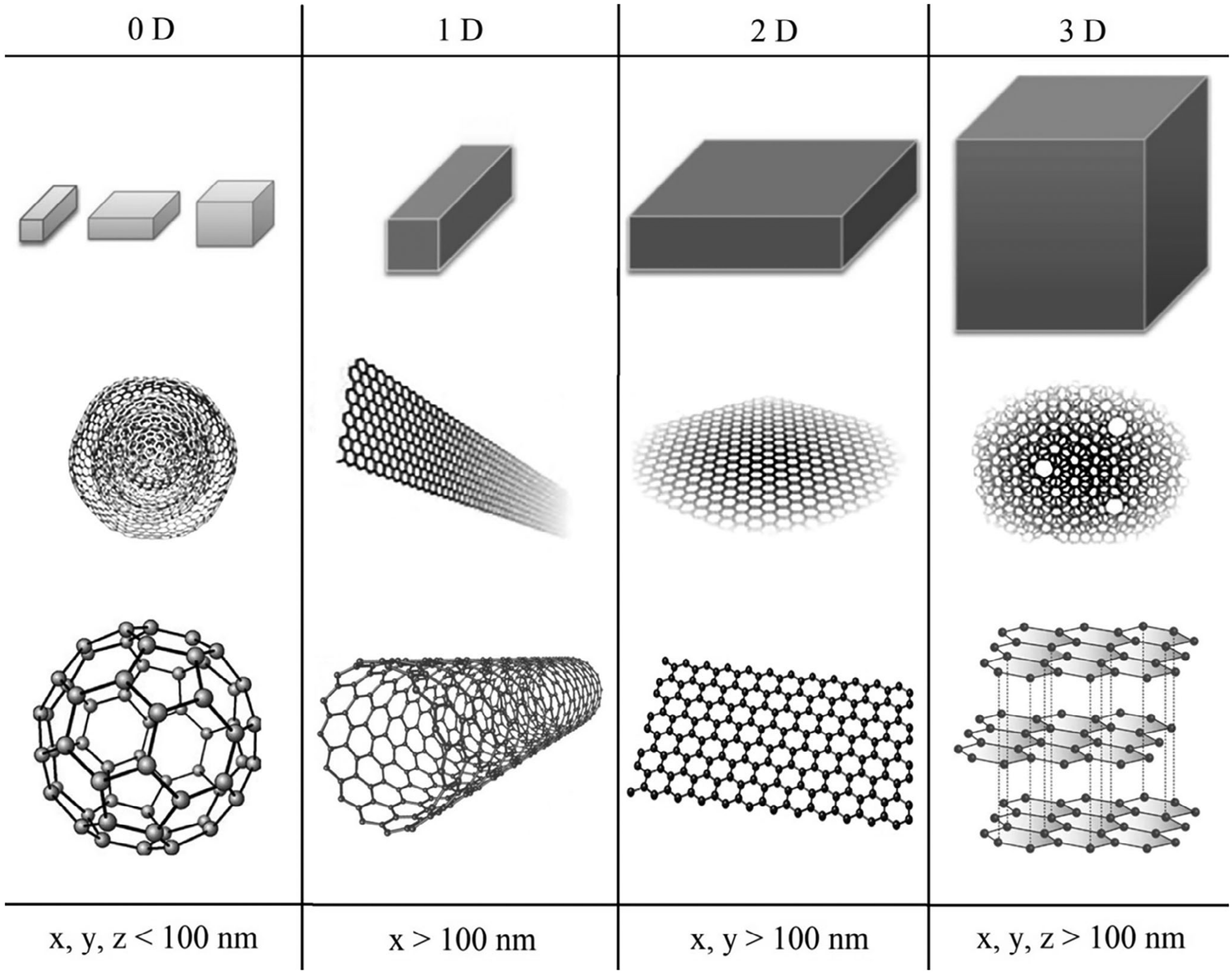

Figure 2. Illustration of different types of dimensional space of a nanostructure.

the size of the nanocrystals should be smaller than the excitation Bohr radius of the material.[7]

The spatial confinement can be in one dimension (1D), two dimensions (2D), or in all the three dimensions (3D) depending upon the confinement of particles in a particular crystallographic direction within a structure and this is illustrated in Figure 2.[8]

So, a typical nanostructure can be classified[9,10] as:

1) Zero dimensional (0D) nanostructure: This materials confined electrons in three dimension or the structure do not permit free particle motion in any direction. Semiconductor quantum dots, nanoparticles, and colloidal particles are some examples of 0D nanostructure.
2) One dimensional (1D) nanostructure: These materials confined electrons in two dimension. Some examples are nanorods, nanowires, nanotube, and nanofilaments, etc.
3) Two dimensional (2D) nanostructure: These materials exhibit a confinement of electrons in one dimension or the structure does not permit free particle motion in one dimension. Examples are nano-discs or platelets, thin-film on a surface, multilayered material, etc.
4) Three dimensional (3D) nanostructure: This is a type of bulk material, which is continuous in three-dimensional space. When system is transited from bulk to quantum dot, the density of states gets reduced gradually.

This paper, in its subsequent section, will discuss about the technologically important CdS semiconductor nanoparticles dealt with the consideration of its synthesis, structural luminescence and magnetic properties, energy interactions and applications.

3. Energy Levels of Rare Earth Ions

Application of luminescent rare earth (RE) ions is mostly obtained by doping in inorganic solids. Basically, there are some important parameters for the mechanism of these display materials like their excitation type and spectrum. Their relaxation to emitting spectra was observed extremely sharp.

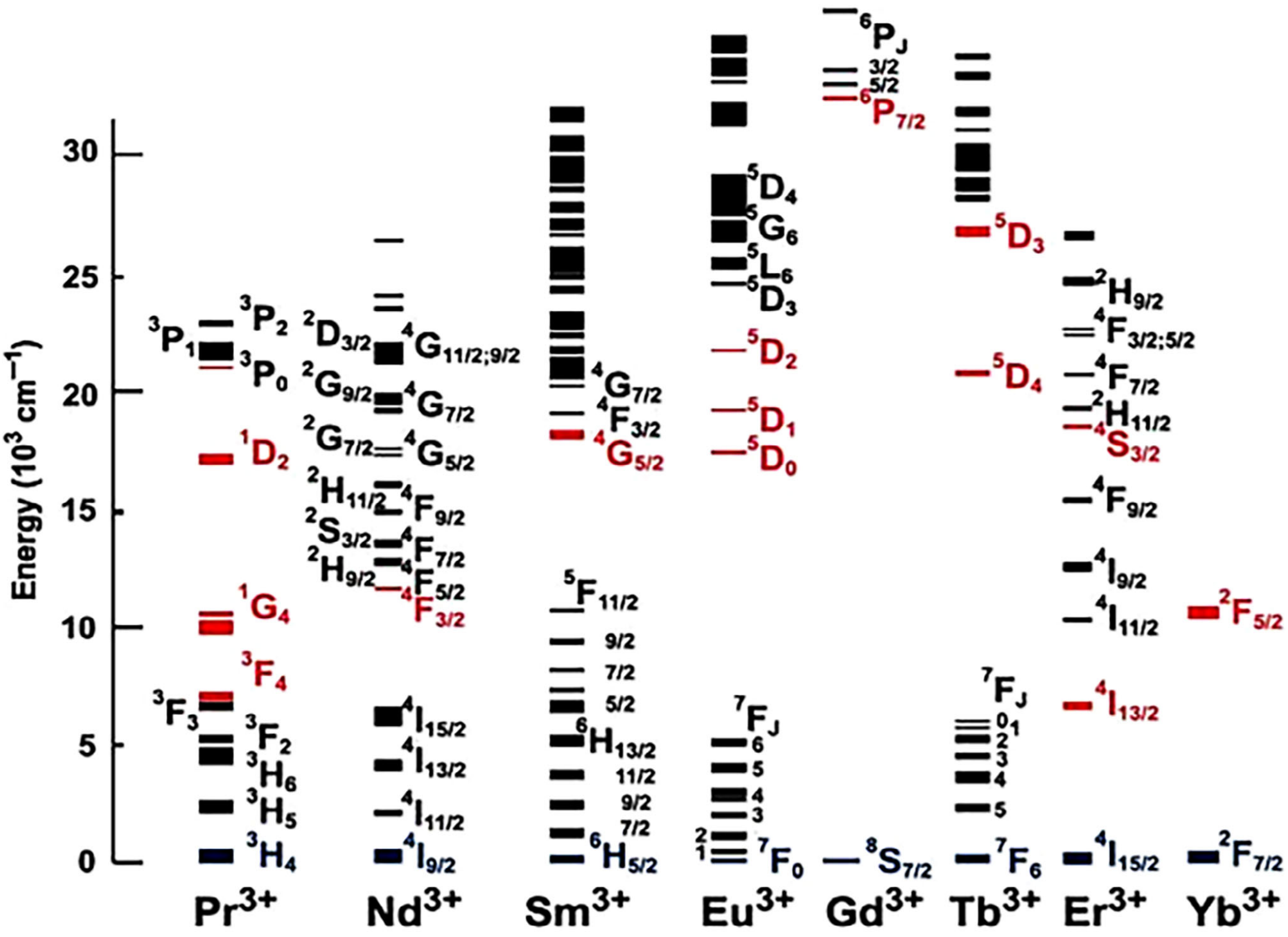

Figure 3. Dieke energy level diagram of RE ions with the main emissive levels highlighted in red.

RE lanthanide ions are characterized by (Xe).$4f^{12}.6s^2$ electronic configuration with the outer shell configuration as $5s^2$, $5p^6$, $6s^2$. The most stable ionization state is the trivalent one, with 5s and 5p electrons acting to screen the energy levels of the 4f electron from the effect of environment.[11] The RE element next to these three tends to exchange electron and get a stable configuration.[12]

To investigate the luminescent properties of RE ions, it is necessary to know their key energy levels. The energy level may be divided into three categories: those corresponding to $4f^n$, $4f^{n-1}$, and 5d configuration. RE ions with crystal field or with the lattice vibration can mix state of different parities into 4f-states. Coupling of 4f electrons with transition dipoles induced in the legends by radiation field leads to an amplification of the even parity multipolar transition amplitudes for transition within 4f shell. These transitions are called as induced electric dipole transition with selection rules $\Delta S = 0$, $L \leq \pm 2$, and $J \leq \pm 2$. These transitions show large variation in oscillator strengths depending upon environment. The transition that is not allowed as electric dipole may take place as magnetic dipole. The magnetic dipole transition obeys the selection rules $\Delta L = 0$, $\Delta J = 0$, and $\Delta J = 1$ $(0 \leftrightarrow 0$ excluded). The Dieke energy level of different RE ions have been shown in Figure 3.

Here we discuss about the praseodymium (Pr^{3+}) and neodymium (Nd^{3+}) ions, which are trivalent RE ions with a complex energy level ${}^1D_2$, ${}^3P_0$, ${}^3P_1$, ${}^3P_2$, ${}^4F_{5/2}$, ${}^2H_{9/2}$, and ${}^4F_{7/2}$, ${}^4S_{3/2}$, ${}^2H_{11/2}$, ${}^4G_{5/2}$, ${}^2G_{7/2}$, ${}^2K_{13/2}$, ${}^4G_{7/2}$, ${}^4G_{9/2}$, ${}^2K_{15/2}$, ${}^2G_{9/2}$ ${}^2D_{3/2}$, ${}^2P_{3/2}$, ${}^4G_{11/2}$ respectively to provide the possibility of simultaneous emissions depending on the CdS materials. Pr^{3+} and Nd^{3+} ion doped fluorescent nanoparticles have been extensively studied because of their properties for application in lighting and display phosphors. It is expected that 4f electrons in the RE ions metal participate in luminescence and ions doped phosphors have emission in the visible range.[13] Pr^{+3} and Nd^{+3} are considered to be most useful among RE ions because they emit light of visible spectrum. Basic aspects of luminescence of the materials as a result of radiative electronic transition in which an electron jump from a higher energy state to lower energy state with the difference in energy released as photons (light emission). For such process to take place the electron must first be excited into a higher energy state by same energy from an external source. The return to the ground state has two paths radiative decay and nonradiatve decay. When an excited electron decays by emitting photons, which is luminescence emission, the process is then called radiative decay. Nonradiative decay is those in which the excited electron decays to the ground state by emitting phonons. Lumi-

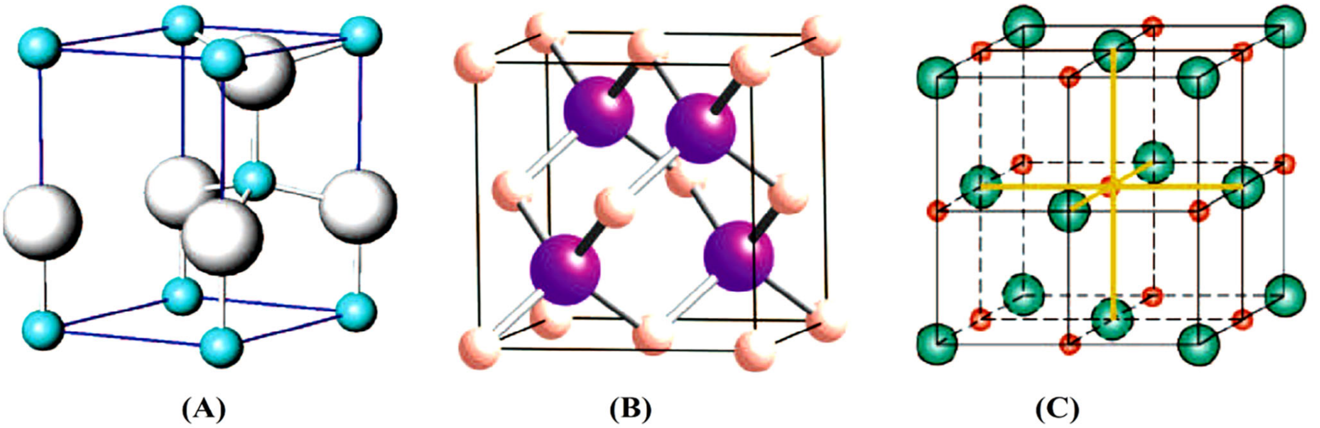

Figure 4. Diagram for the unit cell for crystal structure of CdS: A) Wurtzite (hcp), B) Zinc blend, and C) Rock salt (ccp) phases.

nescence processes begins with absorption of energy by electrons during the 4f-4f transition.[14]

4. Properties of CdS Nanoparticles

CdS nanoparticles can display novel optical, electronic, magnetic, chemical, and structural properties that might find much important technological application.

4.1. Structural Properties

CdS nanoparticles can attain three types of crystal structures namely wurtzite, zinc blend, and rock salt phase. Among this wurtzite is the most stable phase and can be easily synthesized. Wurtzite phase have been observed in both the bulk and nanocrystalline CdS, while zinc blend and rock-salt phases are observed only in nanocrystalline CdS.[15] CdS wurtzite form comprises of hexagonal close packing (hcp). In hexagonal wurtzite and cubic zine blend, each atom is co-ordinated to four other atoms in tetrahedral fashion such that atom has four neighboring atoms of the opposite type. Diagram for the unit cell of the crystal structure of A) wurtzite (hcp), B) zinc blend, and C) cubic closed packed (ccp) rock salt phases of CdS are shown in Figure 4. The melting point, electronic absorption spectra, band gap energy, crystal structure, and other properties of CdS nanoparticles are affected by its size. Thus, CdS on the whole is an attractive system for practicing synthetic chemistry for nanocrystals and for understanding the chemistry, growth history of nanomaterials, and also for technical application.[16,17]

Colloidal dispersions of CdS semiconductor nanoparticles can display color change of fluorescence depending on size of the particle. The CdS nanoparticles show quantum size effect, due to which the size of the CdS particles is directly related to the absorption wavelength. The structure of the nanocrystalline CdS can play an important role in determining the electric properties. It can be crystallized in different structures upon size reduction, depending upon reaction conditions. Physical properties of CdS were collected in Table 2.[17]

Table 2. Physical properties of CdS.

Property	Value
Solubility	Insoluble in hot and cold water
Color	Yellow-orange to brown
Physical state and appearance	Solid (solid power)
Molecular weight	144.46 g mol^{-1}
Melting point (sublimation point)	980 °C/ 1796^{0}F
Specific gravity	4.82 g cm^{-3}

4.2. Preparation Methodology of CdS:Pr^{3+}/CdS:Nd^{3+} Nanoparticles

CdS:Pr^{3+}/CdS:Nd^{3+} nanomaterials were synthesized by simple chemical precipitation synthesis method.[18] This is an easy, inexpensive, and single-step method. The chemicals, used in this work, were of analytical grade without purification. Cadmium nitrate tetrahydrate [Cd $(NO_3)_2 \bullet 4H_2O$], sodium sulphide [Na_2S], diethylene glycol [DEG], ethanol [C_2H_5OH], praseodymium chloride [$PrCl_3$]/neodymium chloride [Nd Cl_3], and distilled water were used as the source material. At first, 50 mL 0.1 M of $Cd(NO_3)_2 \bullet 4H_2O$ solution was taken in conical flask and around 20 mL of DEG was added to this solution with constant stirring. Then, 50 mL Na_2S solution and different concentration (0.1, 0.2, and 0.3 mol%) of $PrCl_3$/$NdCl_3$ were added drop wise and it was kept for reaction for about 4 h at 60 °C with constant stirring. Finally, precipitate of CdS:Pr^{3+}/CdS:Nd^{3+} nanomaterial is formed. It was washed with ethanol and distilled water and dried at 300 K.[19]

4.3. EDAX, SEM, and TEM

Study of the XRD pattern of Pr^{3+} and Nd^{3+} doped CdS has confirmed that the synthesized materials exhibit cubic zinc blend phase of the CdS nanostructure. Peak intensity of these materials decreases with increasing doping concentration. It might be because of some of Pr^{3+} and Nd^{3+} replace the Cd^{2+} ions when increasing the doping concentration. Variation in the nanocrystals has measured from the UV-reflectance spectra. It has also re-

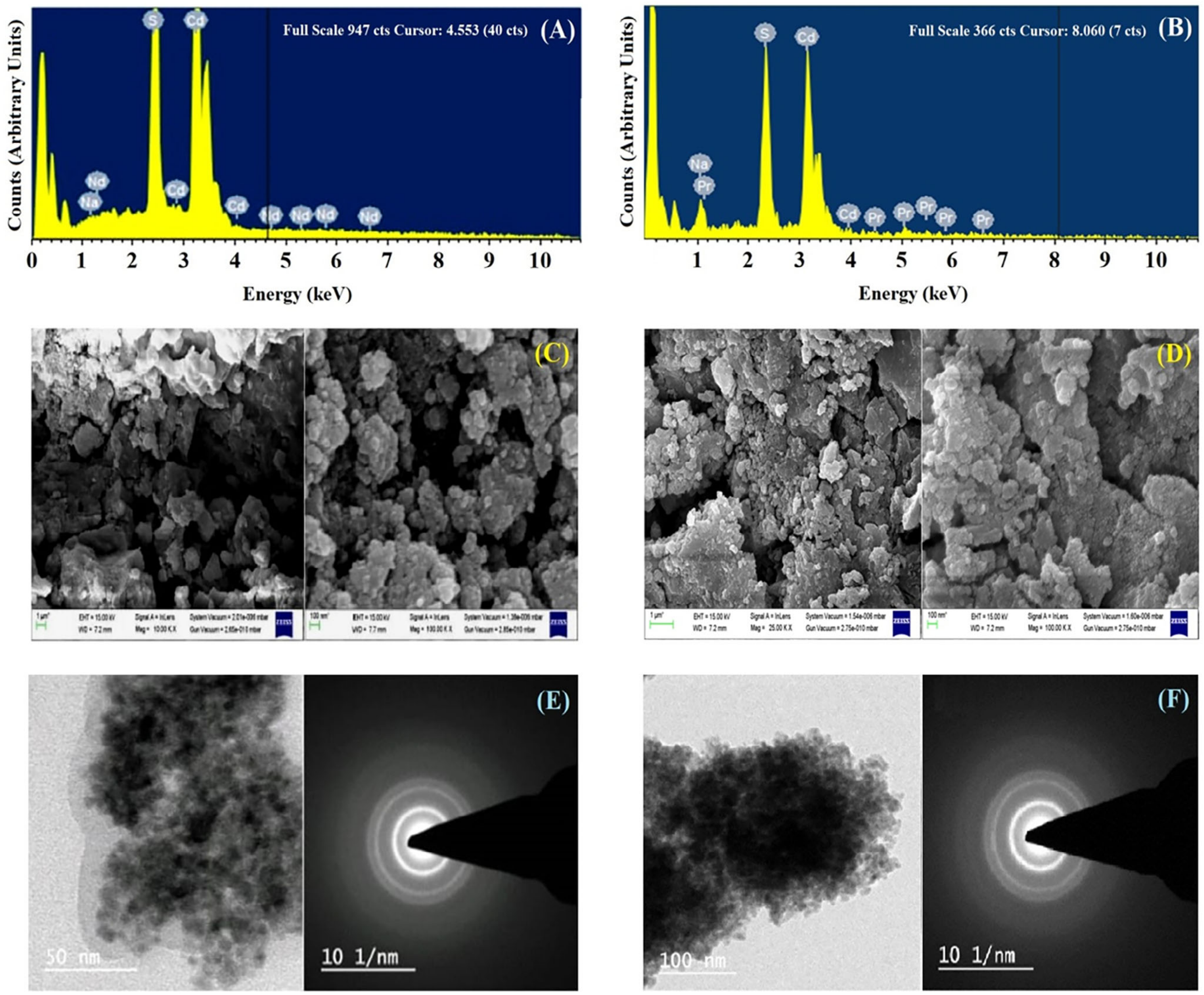

Figure 5. A) EDAX spectrum of CdS nanomaterial with 0.2 mol% doping concentration of Nd^{3+} ion, B) EDAX spectrum of CdS nanomaterial with 0.2 mol% doping concentration of Pr^{3+} ion, C) SEM images of Nd^{3+} doped CdS nanoparticles for 0.2 mol% doping concentration of Nd^{3+} ions at room temperature, D) SEM images of Pr^{3+} doped CdS nanoparticles for 0.2 mol% doping concentration of Pr^{3+} ions at room temperature, E) TEM micrographs (left side) and SAED pattern (right side) of CdS nanoparticles doped with 0.2 mol% of Nd^{3+} ion, and F) TEM micrographs (left side) and SAED pattern (right side) of CdS nanoparticles doped with 0.2 mol% of Pr^{3+} ion.

vealed that increasing dopant concentration decreases the band gap of this structure.[20]

The spontaneous emission probability (luminescence lifetime) of optical transitions from RE ions doped semiconductor nanostructures may be significantly different from their bulk counterparts. In order to improve the luminescence properties of CdS nanoparticles, Nd^{3+} and Pr^{3+} doped fluorescent nanoparticles have been extensively studied because of their properties for application in lighting and display phosphors. It is expected that 4f electrons in the RE ions metal participate in luminescence and ions doped phosphors have emission in the visible range.[21]

Energy dispersive X-ray (EDAX) spectrum of CdS nanomaterial with 0.2 mol% doping concentration of Nd^{3+} and Pr^{3+} ion are respectively shown in Figure 5A,B, SEM images of Nd^{3+} and Pr^{3+} doped CdS nanoparticles for 0.2 mol% doping concentration at room temperature are respectively shown in Figure 5C,D and transmission electron microscopy (TEM) images (left side) and selected area electron diffraction (SAED) pattern (right side) of CdS nanoparticles doped with 0.2 mol% of Nd^{3+} and Pr^{3+} ion are respectively shown in Figure 5E,F. Scanning electron microscopy (SEM) image of these nanostructures reveals spherical shape of the nanocrystals and the size of the particles are around 1 μm–200 nm.

TEM and XRD analysis also reveal the spherical structure of the CdS:Nd^{3+} and CdS:Pr^{3+} nanoparticles in the prepared samples. Spectroscopic analysis of trivalent Pr^{+3} and Nd^{+3} ions have received very much interest and reveals good fluorescence efficiency in visible and near IR (NIR) region. Doping of Pr^{3+} and Nd^{3+} with TiO_2 was beneficial for NO_2 absorption to enhance the photocatalytic activity.[22] The impact of RE ions on the structural, optical, dielectric, and magnetic properties of copper nanoferrites has been investigated through the various characterization facilities to explore the feasibility for application in magnetic devices.[23,24]

Pr^{3+} doped CdS quantum dot silicate glass can be synthesized by melting quenching method. The crystallization behavior and luminescent property of the Pr^{3+} doped CdS silicate glass were investigated by characterization TEM, SEM, X-ray diffraction (XRD), UV-NIR, and fluorescence spectra. It was found that with increasing heat temperature, the size of CdS particles increased from 2.30 to 3.63 nm.

Luminescence intensities are observed in glass containing Pr^{3+} doped CdS quantum dot.[25,26] Doping of CdS nanoparticles Nd^{+3} ions, results in an emission at 877 nm under 405 nm excitation due to transfer of energy from the CdS nanoparticles to Nd ions. Analyzing the dynamical luminescence of the $^4F_{3/2}$ state, we also observed that the Nd ions are located in two sites, embedded in the glass matrix and on the CdS nanoparticles.[27]

The UV-visible spectra of $NdCl_3$ and $PrCl_3$ were successfully obtained in LiCl-KCl eutectic at 450 °C. Both elements follow the Beer Lambert law at least up to 1.5 wt% for $NdCl_2$ and 3 wt% for $PrCl_3$. The molar absorption coefficients were successfully calculated for the different peaks. The precipitation of each component by oxygen addition was successfully monitored by UV-visible spectroscopy.[28] The preparation of Pr^{+3}, Nd^{+3}, doped CdS nanoparticles was done by using simple aqueous precipitation method at room temperature.[29]

5. Applications

CdS nanoparticles show very sharp luminescence lines which hardly depend on temperature and crystal host. CdS belongs to II–VI group and is a well-known semiconductor having direct band gap of 2.42 eV at room temperature. This enables its nanoparticles to be remarkable in optoelectronics, photonics, and photovoltaics. Doping with transition metal elements into CdS nanoparticles leads to many interests and good structural, electrical, and optical properties have been reported. Chemical precipitation method is one of the most important techniques used in industrial applications. The polymers may be good stabilizers as they can interact with the metal ions.[30–32]

CdS nanoparticle amplified the luminescence intensity of Pr^{+3}, Nd^{+3} ions by up conversion processes. So energy transfer can be observed from CdS nanoparticles doped with Pr^{+3}, Nd^{+3} ions. Therefore, the use of pump sources and diode lasers in the UV range can be used to excite these samples to work as lasers.[33]

RE doped nanocrystals show a broadband emission in the visible region when excited by high-energy photons below the band edge of host CdS nanocrystals.[34] The structure of CdS possesses intensity optical properties due to their good interaction with light and provides significant advantage to semiconductor which is important for photoluminescence and other optical properties.[35–37] The element structure of RE elements is the same at their outer shell ($5s^2$, $5p^6$, $6s^2$) and differs only by electrons in the partially filled inner 4f shell.

RE ion doped CdS nanoparticles find potential application as a window material for hetero-junction solar cell light emitting diodes (LED), photo-catalysis, biological sensors, address decoders, gas detectors, etc. Transistor metal ions and RE ions have been doped into CdS nanostructures which act as recombination centers for the excited electron-holes pair and give out tunable emission.[38] CdS nanomaterials are also used as pigment in paints and in engineered plastic due to their good thermal stability.[39] CdS nanoparticles can be used in optoelectronics for marking photocells, LED,[40] lasers, field effect transistor [FET],[41] etc. In photonics, due to its photo-conducting and electrical properties it can be used in sensers, photodetectors, optical filters, and optical switches. As its band gap appears in the visible spectrum,[42] it is useful for many commercial and potential applications in photovoltaics, as hetero-junction solar cells and thin film solar cells. In photocatalysis, owing to its photochemicals and catalytic poperties, CdS nanoparticles can be used for water splitting[43,44] as well as for water and air purification. Among water splitting photocatalysts, CdS is considered to be an interesting and important material owing to its low cost, prominent catalytic activity, high absorption in the visible spectrum, and the suitable positions of its conduction and valence bands.[45] CdS nanomaterial can be used for the diagnosis and treatment of cancer due to its high optical and fluorescence properties. Diagnosis or imaging of cancer cells can be done by accumulating CdS nanoparticle inside cancer cells, which then can be easily visualized by irradiated with ultraviolet (UV) radiation for treatment of cancer. Photo activation of fluorescent CdS nanoparticle (photodynamics cancer therapy) accumulated with in cancer cell with radio sensitizing agents could induce cell death. CdS nanoparticle can be used for the purposes of visualization as well as for drug delivery to tissues of the eye including retina and cornea.

On the other hand, the quantum dots or nanocrystals have interest due to distinct properties from that of bulk materials as a wide absorption spectrum compared to RE. The state was confirmed through emission spectra, which also indicate a decrease in the number of defects with increase of the nanocrystal size. The quantum dots have the potential to improve photoelectric conversion efficiency of solar cells for the change of Fermi level band gap and conductance. The structure and properties to the electron delivery and collection led to an improved efficiency. The application of RE materials doping in dye sensitized solar cells is a new exploration. Up-down conversion by doping with RE ions can cause optical luminescence.[33,46,47]

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

Research data are not shared.

Keywords

absorption spectrum, CdS nanomaterial, energy parameters

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