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Review Article

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Nanotechnology of colour: quantum dots (QDs), photonic crystals (PCs) and plasmonic nanoparticles

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Abstract: This tutorial min review discusses the important features of nanotechnology of colour generation by the quantum dots (QDs), nanostructured photonic crystals (PCs) and plasmonic metal nanoparticles. Quantum dots (QDs) are characterized by the tunable and size dependent strong fluorescence emission properties. Photonic crystals (PCs) generate the material independent structural colour (***iridescent colour dependent on the angle of observation***) due to the constructive interference arising from Bragg diffraction by the nanostructured photonic crystals (PCs) producing the diffraction gratings. It causes the vibrant colour in the opals and pearls, wings of peacocks and butterflies, in the necks of pigeons, variable colours in chameleons, etc. Plasmonic nanoparticles like Au-, Ag- and Cu-nanoparticles show the both light absorption and scattering properties that can be tuned by varying the size, morphology and composition of the nanoparticles. Structural colour (***iridescent colour***) by PCs is used by nature while colouring by QDs and plasmonic nanoparticles are due to the man made activities. The nanotechnology based colour is superior to the organic pigment based colour in terms of brightness and photostability and this field is an emerging area of research with a great promise in terms of applications in various fields.

Keywords: quantum confinement (QC); quantum dots (QDs); photonic crystals (PCs); plasmonic nanoparticles; Lycurgus cup; dichoric property

1 Introduction

The nanotechnology based colour is mechanistically different from the chromophore based colour. The properties of nanomaterial are now well understood and it is still an active area of research from the stand point of their applications in different fields including the medical field. The semiconductor quantum dots (QDs) are the man made materials.[1,5,24,59] They have the tunabe and size dependent fluorescence emission properties that may find many potential applications in colour display, bioimaging, light emitting diodes (LEDs) and in many other optoelectronic devices. QDs are superior to the conventional organic fluorescent dyes in terms of intensity and photostability. The material independent photonic structure colour due to Bragg diffraction from the diffraction grating produced by the nanostructured PCs has been used by nature to generate the ***vibrant iridescent colours*** as in the opals and pearls, wings of peacocks and butterflies, in the necks of pigeons, variable colours in chameleons, etc. The plasmonic nanoparticles ***like Au-, Ag- and Cu-nanoparticles*** showing the tunable optical properties due to the localized surface plasmon resonance **(LSPR)** are the man made products like the QDs. The use of plasmonic nanoparticles in colouring the ceramics and glasses was known to the glass makers and ceramic artists in the ancient days but science behind this colouring nanotechnology is explored in the recent days. The colouring due to the photonic crystals and plasmonic nanopartices can survive for a long time while the colouring by the chromophore based organic pigments becomes faded on standing and exposure to environmental factors. This is why, the active research on the possibility of the use of photonic crystals and plasmonic nanoparticles in colouring different objects is going on. This article aims to summarise the various aspects of the ***nanotechnology of colour generation*** which is not generally discussed in a single source.

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2 Colourful fluoresence emission from the semiconductor quantum dots (QDs): a nanotechnology of colour

2.1 Quantum dots (QDs) and effect of quantum confinement (QC) on the optical properties of QDs

Quantum dots (QDs) are the 0-D (zero dimensional) nanoparticles of a semiconducting material like **12–16 type semiconductor** (CdX, X = S, Se, Te) and **13–15 type semiconductor** (InP, GaAs).[2,15,59] QDs experience the **quantum confinement** (QC) in all directions.[3,4,9,33,39,52] The condition of QC for the electrons in the QD (quantum box) is that the radius of the semiconductor nanocrystal is to be smaller than the **exciton Bohr radius**.[15,59] The exciton is the electron-hole pair (*i.e.* excited electron in the conduction band and the hole it leaves behind in the valence band). The exciton Bohr radius is the average distance between the electron in the conduction band (CB) and the hole in the valence band (VB). The exciton (electron-hole pair, called **Wannier-Mott exciton**) may be considered to constitute an H-atom in the solid state of semiconductor where the hole of positive charge in the VB replaces the hydrogen nucleus and the electron in the CB replaces the H-electron.[15,59] In inorganic semiconductors, the dielectric constant is generally large (*ca.* 10–40 while it is taken as unity in air) due to the presence of a large number of electrons surrounding the exciton (*cf.* H-atom: 1 electron + 1 proton system). This high dielectric constant reduces the Coulombic attractive force in the exciton. Besides this, the **effective reduced mass** of an exciton is also small. These two factors (*i.e.* higher dielectric constant and smaller reduced mass) make the exciton binding energy smaller and radius much larger than those of H-atoms (*e.g.* for GaAs semiconductor, $E_{\text{exciton}} = 4.6$ meV, $r_{\text{exciton}} = 11.5$ nm while $E_{\text{H}} = 13.6$ eV and $r_{\text{H}} = 0.53$ nm).

Due to the QC in all directions in QDs, they possess the **discrete and quantised energy levels** instead of the continuum of energy levels of the bulk semiconductor (Figure 1). Thus in terms of the energy levels, they are more closely related to the free atoms than to the bulk materials. This is why, the QDs are nicknamed as the **artificial atoms or superatoms**.[3,24] Due to the QC in QDs, the band gap (E_g) between the valence band (VB) or HOMO band (consisting of discrete energy levels) and conduction band (CB) or LUMO band increases compared to that of bulk semiconductor. The band gap dependence on the size of QDs can be expressed as follows by the Brus equation given below.[4,9]

$$E_{\text{g}}^{\text{QD}} = E_{\text{g}}^{\text{Bulk}} + \frac{h^2}{8R^2}\left(\frac{1}{m_e^*} + \frac{1}{m_h^*}\right) - \frac{1.8e^2}{\varepsilon_0\varepsilon_r R}$$

$$= E_{\text{g}}^{\text{Bulk}} + \underbrace{\frac{h^2}{8R^2\mu}}_{\substack{\text{Causing}\\ \textbf{(Blue Shift)}}} - \underbrace{\frac{1.8e^2}{\varepsilon_0\varepsilon_r R}}_{\substack{\text{Causing}\\ \textbf{(Red Shift)}}}$$

where $\varepsilon = \varepsilon_0\varepsilon_r$, E_g^{Bulk} = band gap in bulk (the difference in energy between the maximum of the VB and the minimum of the CB), E_g^{QD} = band gap in the spherical nanoparticle or quatum dot having the radius R, and μ = reduced mass of the system consisting of effective mass of the hole (m_h*) and electron (m_e*).

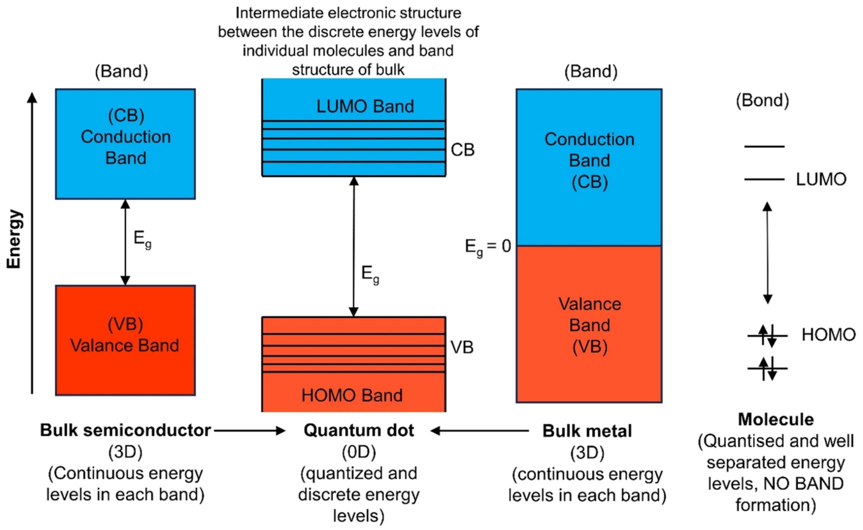

Figure 1: Comparison of the energy levels in the bulk semiconductor, bulk metal, quantum dot (QD) and an individual molecule. Continuum of energy levels in the bulk 3D state is replaced by a set of quantized discrete energy levels in 0D QD.

The second term of Brus equation represents the **quantum confinement energy** (*cf.* ‘*particle in a box model*’) and this is responsible for **blue-shift** while the third term represents the **Coulombic interaction energy** and is responsible for **red-shift**. The quantum confinement energy term (responsible for blue-shift) having the size dependence given by R^{-2} is more sensitive to the variation of size (R) of the nanoparticle compared to the Coulombic interaction energy term (responsible for red-shift) having the R^{-1} dependence when R is small (approximately few nm). In fact, because of these different types of dependence on R and high dielectric constant (ε) of the material, the Brus equation can be reasonably written as follows.[15]

$$E_g^{QD} \approx E_g^{Bulk} + \frac{h^2}{8R^2\mu}\,;\ i.e. R = \sqrt{\frac{h^2}{8\left(E_g^{QD} - E_g^{Bulk}\right)\mu}}$$

It indicates that the band gap energy in QDs of a particular composition can be tuned by controlling the size of the particle. The band gap energy determines the wavelength of **band-edge fluorescence emission** (Figure 2). When the incident light excites an electron from the VB to the CB of a QD, it creates an exciton (electron–hole pair). The promoted electron in the CB quickly relaxes nonradiatively to the lowest CB edge while the hole in the VB moves towards the topmost level of VB (Figure 2). This rapid nonradiative thermal relaxation places the electron and the hole at their most stable positions, *i.e.* band edges of the respective bands. Thus the stable exciton is generated by placing the excited electron at the lowest CB edge and the hole at the highest VB edge. They recombine in a radiative path giving the band edge fluorescent emission that gives the monochromatic emission (*i.e.* sharp emission band).

According to Brus equation, the wavelength of band edge fluorescence emission of QDs can be tuned by changing their size: ***red shift of*** λ_{max} ***with the increase of particle size*** (Figure 3). ***The smaller QDs give the fluorescence emission of shorter wavelengths to produce colors such as violet, blue or green while the bigger QDs give the fluorescence emission of longer wavelengths to produce colors like yellow, orange or red***. This tunable and **size-dependent optical properties of QDs** make them the promising alternatives to the organic lumophores. They show the unique optical properties:[36,42,43,59] *wide absorption (excitation) band extending towards the UV region and narrow but strong fluorescence emission band (i.e.* narrow and symmetrical emission band at specific wavelengths), *high quantum yield, high photostability, and high molar extinction coefficients for both excitation and emission.* The intensity of their fluorescence emission is much stronger than that of organic fluorophores. In fact, the colour of the fluorescence emission of QDs can be tuned in the whole visible spectrum extending to the near-infrared (NIR) range (700–900 nm) which represents the **biological transparent window**. Such NIR QDs are important in cellular imaging because the NIR fluoresecent emissions are not absorbed by the blood and tissues. In fact, QDs may find versatile applications in colourful display and bioimaging and nanoelectronics.[1,5,32] This emerging field with a great promise has been recognized by the **Nobel Prize in Chemistry 2023** to M. G. Bawendi, L. E. Brus and A. I. Ekimov for the discovery, synthesis and development of QDs.

2.2 High intensity of QD fluorescence emission

It is due to the following reasons.[15,36,42]

- **High transition probability:** According to the condition of electron-photon momentum matching, optical transitions in the **indirect band-gap semiconductors** (where the band edges are at different *k*-values in the

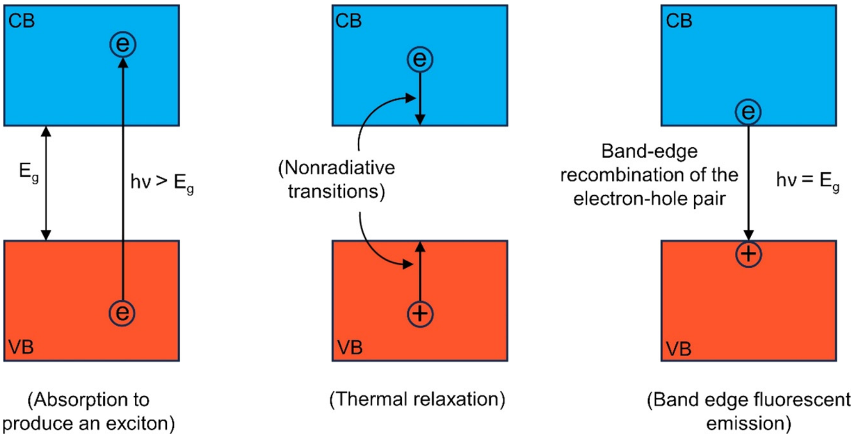

Figure 2: Qualitative illustration of photoexcitation to generate the exciton, thermal relaxation of the electron and hole of the exciton followed by the band edge fluorescent emission in QD.

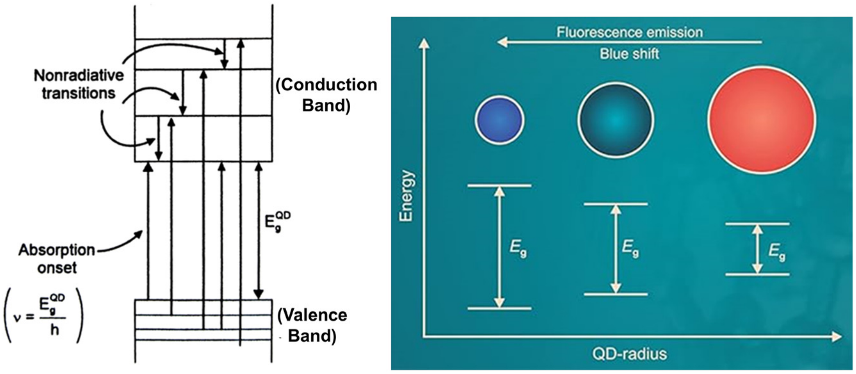

Figure 3: Illustration of quantum confinement effect on band gap (E_g) and optical transitions in quantum dots. (a) Qualitative representation of the energy levels of semiconductor quantum dots (QDs) showing the optical excitations. Both VB and CB consisting of discrete energy levels due to quantum confinement (QC) effect. **Band-edge fluorescence emission** from the bottom energy level of CB to the topmost energy level of VB. (b) Qualitative representation of the dependence of band gap (E_g) and band-edge fluorescence emission on the size of QDs (Brus equation). **Absorption onset** indicates the lowest energy end of the broad absorption band for excitation.

E–*k* plot, *k* is called the wave vector which is proportional to the momentum of the electron) are **momentum-forbidden** and it needs the assistance of lattice phonons to generate the electron-hole pairs but this problem does not arise for the **direct band-gap semiconductors** (where the band edges are at the same *k*-values in the *E*–*k* plot). In quantum dots, the momentum matching condition is not applicable because under the quantum confinement (QC) effect, the electrons are not associated with the linear momentum vector (*i.e.* no net linear momentum in terms of the 'particle in a box' model).[15,59] Thus the transition probability in both the radiative excitation and fluorescent emission is very high.

- **Prevention of the nonradiative exciton decay across the band gap from the stable position of exciton**: For this interband nonradiative transition path, the energy change is relatively high and the nanocrystal cannot accommodate such a large thermal vibration without breaking the crystal. It eventually prevents this nonradiative decay path and favours the band edge fluorescent emission.
- **Favourable and noncritical excitation characterized by a broad absorption band:** The exciting radiation frequency can cover a wide range starting from the **absorption onset** frequency (minimum value determined by the E_g value, $E_g = h\nu_{\text{min}}$) extending towards the UV region (*i.e.* a broad absorption band). On the other hand, for an organic fluorescent dye (*e.g.* **fluorescein isothiocyanate**, FITC) having no energy band structure, the absorbance and emission occur almost at the same wavelength.
- **Prevention of red-shifted defect fluorescent emission through surface passivation**: The trapped electrons and holes by the **surface dangling orbitals and surface defects** may undergo the radiative combination to give the red shifted defect fluorescent emission to weaken the intensity of band edge fluorescent emission. But this can be avoided through the surface passivation.
- **Better optical properties of core–shell semiconductor QDs** (*e.g.* CdS@CdSe, CdSe shell and CdS at the core): The surface atoms of a QD being coordinatively unsaturated possess the dangling or unpassivated orbitals (*i.e.* unsatisfied valence orbitals). The electrons and holes may be trapped by these dangling orbitals. Besides this, the surface defects can also trap the electrons and holes. The shell of nanometer-scale thickness can passivate the core to improve the optical properties of the core. The shell also protects the core from the environment and photo-oxidative degradation.

2.3 Applications of optical properties of quantum dots (QDs)

The tunable optical properties of QDs find many important applications.[1,5,15,32,42,59] These are discussed here.

- **Bioimaging:** It is important to the diagnoses of many diseases including cancer cells, to track the target biomolecule, and to understand the different aspects of immunology. For bioimaging, QDs are superior to the traditional fluorescent organic dyes in terms of quantum yield (brightness) and photostability. For bioimaging by the QDs, they are to satisfy the conditions: fluorescent emission in the NIR range (biologically transparent window) and to remain well-dispersed and

stable in the aqueous medium in the physiological pH range. Now different methods have been developed to synthesise the water-dispersible NIR QDs. The limitation of metal based QDs like CdX (X = S, Se and Te) is due to their toxicity. The nontoxic Si-QDs may be considered as the alternatives discussed later.
- **Light emitting devices:** QDs find a potential application in light-emitting diodes (LEDs). In fact, QDs are very useful for the colourful display devices because of their tunable bright fluorescent emission in the entire visible range spectrum.
- **Photovoltaic devices:** QDs are now being considered as the potential candidates for light energy harvesting photovoltaic cells because of their tunable absorption spectrum matching with the UV–visible range and high extinction coefficient. In fact, Si-based photovoltaic cells are highly costly and its alternative cost-effective devices are of much importance.
- **Photocatalysis**: QDs can also act as the photocatalysts in a similar manner of photocatalysis by the bulk semiconductor like TiO_2. The advantage of QDs is that its bangap can be tuned according to the requirement.

2.4 Silicon quantum dots (Si-QDs): a new generation nontoxic QD

The metal based QDs very often possess the toxic elements like Cd, As, Se, etc. This is a limitation of such QDs. Si-QDs being nontoxic are the promising alternatives to the metal based QDs.[34,50,55,59] Si-QDs are the semiconductor Si-nanoparticles (size: 1–10 nm). Silicon in the bulk state is an **indirect band gap semiconductor** but in the nano-range (particle size <5 nm), it appears as a **direct band gap material** with a high quantum yield of photoluminescence (PL). Si-QDs have also the **tunable optical properties** that can be controlled by varying the particle size (*i.e.* blue shift with the decrease of particle size in fluorescence emission, **Brus equation**), the nature of the surface groups, and by doping with the transition metals. Si-QDs find versatile applications in optoelectronic devices including the LEDs (Light emitting diodes) and fluorescent biological imaging due to their tunable optical properties and nontoxicity. Si-QD synthesis can be done by both the top-down methods (*e.g.* laser ablation or electrochemical etching of Si-wafer), and bottom-up methods (*e.g.* reduction of silicon halides by sodium, lithium aluminum hydride). The **nontoxic silicon quantum dots (Si-QDs)** are the new generation QDs as the promising alternatives to the toxic metal based QDs but this field needs more studies.

3 Iridescent colours and structure colour: nature’s nanotechnology of colour and photonic crystals (PCs) (nanostructure)

3.1 Photonic crystals (PCs): origin of iridescence and iridescent colours

Before to discuss the photonic crystals (nanostructure) and structure colour, we should understand the meaning of iridescence and iridescent colours. Iridescent colours are characterized by the change in colour with the direction of observation and it is caused by optical interference arising from the incident light on a diffraction grating (**Bragg’s law**). **Photonic crystals (PCs)** represent the *self-assembled nanostructures having the dimension comparable to the wavelength of visible light with the ordered periodic arrangements to form a diffraction grating*. PCs produce the dielectric multilayer structures to act as the diffraction gratings. The incident light on such a diffraction grating may experience both the constructive and destructive interference depending on the wavelength of the incident light and angle of incidence (Bragg’s law, condition for constructive interference: $n\lambda = 2d\sin\theta$ where d = interplanar distance, *i.e.* vertical distance between the successive diffracting planes, n = order of diffraction = 1, 2, 3, … and θ = angle of incident light). **The constructive interference leads to a strong reflection of light of a specific wavelength to determine the observed colour of the object having the nanostructure diffraction grating**.[46,57,59] This selective strong reflection **(called iridescence)** may be compared with the selective absorption by the chromophore of pigments for which we see the complementary colour which is not absorbed.

To understand the origin of iridescence due to the constructive interference, let us first explain the well known **iridescent colours (play of colours in a sunny afternoon) of soap bubbles** having only two surfaces (*i.e.* outer and inner surfaces) for reflection of the incident light where the thickness separating the two reflecting surfaces is comparable to the wavelength of the visible light (≈380–700 nm). Light waves reflected by both the outer and inner surfaces of the thin layer of the soap water bubble experience the **constructive interference,** if the reflected waves are in the same phase and **destructive interference** (*i.e.* cancelling out each other), if the reflected waves are in the opposite phase. Let us consider that white sun light is incident at a specific angle on a soap bubble whose layer thickness is such that only the incident green (G) light experiences the cancelation effect (*i.e.* destructive interference). In this case, both red (R) and blue (B) lights are reflected and then the soap bubble looks magenta (=R + B) coloured (*cf.* **RGB model of colour**). Due to gravity, the thin layer of a soap water bubble becomes thicker at the bottom surface and thinner at the top surface. Thus we see the different colours from the different parts of the soap bubble. The observed colour also depends on the angle of observation.

3.2 Photonic bandgap (PBG) in photonic crystals (PCs)

We shall discuss the PBG in a qualitative way without the mathematical treatment. Photonic crystals (PCs) are constituted by the self-assembled monodispersed nanostructures with the periodic variations in the dielectric permittivity (*i.e.* periodic arrangement of dielectric materials with different refractive indices). 1D, 2D and 3D PCs consisting of the periodic arrangement of dielectric materials are known. Due to the Bragg diffraction caused by the periodic structure of photonic crystals (PCs), some of the incident electromagnetic waves will be hindered to propagate through the photonic crystal structure and form the photonic bandgap (PBG), a band of frequencies for which light propagation is not allowed through a particular PC forming a diffraction grating. In fact, PBGs of PCs can control the propagation of photons and this property can be compared with the *electronic bandgap of semiconductors to control the flow of electrons*. The property of PBG to prevent the propagation of photons of some specific wavelengths can be utilised in many applications like PC diodes and transistors in all optical computers, PC lasers, LEDs, optical fibres, etc.

Electromagnetic waves whose frequencies are outside the PBG will propagate through the PC smoothly. **The electromagnetic waves within the PBG will be prevented from spreading within the periodic array of the PCs and reflected back due to Bragg diffraction to produce the bright structural colours**. By adjusting the size of the PCs, lattice spacing (lattice constants) of the PCs or the refractive index of the photonic crystal material, the PBGs of the PCs can be tuned in the visible range to give different photonic crystal structural colours.

3.3 Photonic crystals (PCs) and structural colour in nature (nature's nanotechnology of colour)

Nature has developed the PCs (*self-assembled nanostructures forming a diffraction grating to the incident visible light*) to give the brilliant iridescent colours like *colour play of pearls, opals, peacock's feather, pigeon's neck, chameleons, butterfly, beetle, different birds and insects*, etc.[6,12,41,49,51,53,59] In contrast to the pigment based colours (*e.g.* melanin, carotenoid, anthocyanin, porphyrin, etc.), the *photonic crystal structural colour is much brighter and size dependent, periodicity (periodic microstructure) dependent and refractive index dependent.* Pigments are photobleached but PC structural colour is never bleached and these biological colours survive for a long time even after death of the corresponding living species. In fact, many living species use the PC structural colour for camouflage (protection) and mating display (attracting partner). Because of these advantages, *possibility of textile colouring by using PCs and replacement of pigment based colour in different industrial fields are being actively explored.* Let us illustrate some specific photonic crystal structural colours in nature.[30,56,59]

- **Opal (gemstone)**:[38,48] This sedimentary gemstone does not have a crystalline structure, but it consists of *nanospheres of silica assembled together.* **Blue opal** (*i.e.* selective reflection of bluish light) is well known. In fact, **opal** displays a **play-of-colour as in other structural colours.** In fact, it is an example of a naturally occurring PC consisting of silica nanospheres arranged/packed in an ordered way to produce a diffraction grating for the visible light (spacing between the silica nanospheres is approximately 150–300 nm, and approximately comparable to the wavelength of the visible light). The observed spectacular colours (different in different angles) called **opalasence** is due to Bragg diffraction from the stacked lattice planes constituted by the silica nanospheres (constructive interference to give a bright reflection of a particular colour depending on the angle of incidence, as explained already). The observed PC structural colour of opal depends on the size of the silica nanospheres and their periodicity.
- **Pearls**:[37] These colourful fascinating gems are produced by oysters and other mollusks that live in seas or lakes. Depending on the type of oyster and nature of water of lakes or seas, pearl colours can be white, silvery white, pink, golden, cream (a light yellowish brown), gray or a combination of these producing attractive shades. The bright luster and **iridescent colours** of pearls are the PC structural colours, which originate from multiple Bragg reflections in a ***nanolayered structure of nacre*** (known as mother of pearl). Nacre is composed of **nanosized aragonite** (one polymorph of $CaCO_3$ crystal, other two polymorphs are calcite and vaterite) crystal (PC) layers separated by conchiolin, a protein produced by mollusks and oysters. This **nanocomposite material** produces their shells for protection. The thickness

(*i.e.* lattice constants) of these multiple layers determines the colour of reflection from the nacre. The colour also depends on the angle of observation.

Here it may noted that bone and nacre are the unique examples of **natural nanocomposites** to introduce the structural toughness. In both these natural nanocompsites, inorganic (ceramic) nanomaterials are dispersed in an ordered way into the organic matrix. In bone, *hydroxyapatitte nanocrystals* are dispersed in the collagen polymer matrix.[15]

- **Chameleons** (a group of lizard species):[47] They can change and display different colours for different purposes like camouflage and social communication. Display of red, orange, or yellow skin colour indicates the sign of excited states like aggression towards another chameleon or animal. The blue and green colours are displayed to indicate calmness, relaxed state or submission to an aggressor (Figure 4). The nano-sized crystals of guanine form the PCs on the skin of chameleons. The rearrangements of these guanine nanocrystals (PCs) on their skins are different in their relaxed and excited states (Figure 5). This arrangement patterns of guanine PCs on their skins can be changed by skin contraction and expansion. This rearrangement of PCs changes the lattice spacings (lattice constants) of the reflecting planes to produce different diffraction gratings and causes structural colour change in these chameleons.
- **Peacock's feathers (plumage, a layer of feathers)**:[20,58] The PCs made of melanin nanocrystals connected by the keratin protein are responsible for the structure colour of peacocks. Orientation of these PCs can give the four principal colours blue, green, yellow, and brown on the peacock feathers. The lattice constants of these PCs are different in these different coloured barbules to give the different diffraction gratings to give the different colours. These colourful feathers are not found in females who are to protect the youngs.
- **Iridescent colours of pigeon's neck and feathers**:[40] The iridescent feathers are more common in male pigeons as in male peacocks. The colour of these feathers depends on the angle of observation, a characteristic property of iridescent colour (PC structural colour). In fact, if a pigeon turns its neck, its iridescent feathers can shift the appearance from purple to green, or vice versa.
- **Iridescent wings of damselflies**:[10,45] The colourful properties of their wings arise due to the nanostructural components. The nanostructured layers acting as the diffraction gratings in their wings are composed of different chemical components like chitin (polysaccharide, a long-chain polymer). This nanoperiodicity makes the wings to appear iridescent which is important for their camouflage (protection) and reproduction.

3.4 Photonic crystal structural colour in industrial application

The photonic crystal structural colour is not photobleached while the organic chromophore based colour is photobleached on standing in the environment. This is why, the possibility of colouring of many industrial products including the textiles by using the photonic crystals has been explored.[30,56]

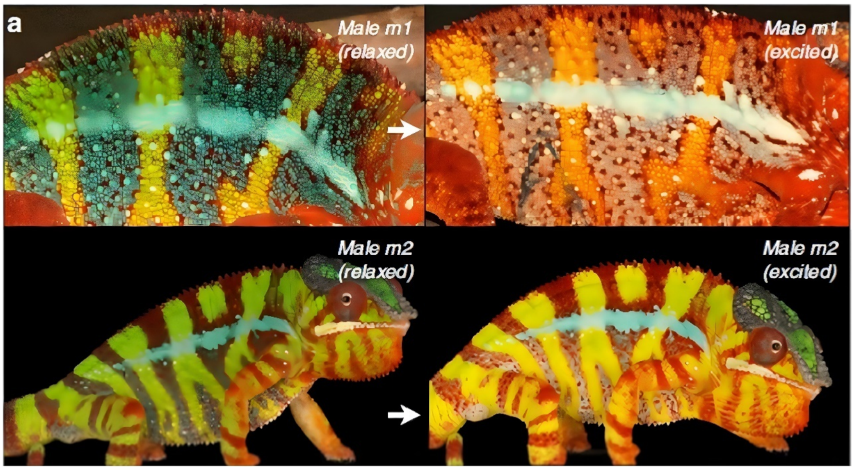

Figure 4: Left two images for the relaxed chameleons and right two images for excited chameleons as in male-male combat (image by Teyssier[47]; courtesy of CC BY 4.0).

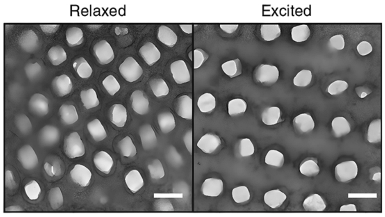

Figure 5: Microscope images of chameleon skin in the 'relaxed state' (left side) and in the 'excited state' (right side) showing the difference in lattice constants of photonic crystals (PCs) forming the diffraction gratings. Scale bars = 200 nm (image by Teyssier[47]; courtesy of CC BY 4.0).

4 Colours of the plasmonic metallic nanostructure: a nanotechnology of colour

4.1 Surface plasmon resonance (SPR) of plasmonic metallic nanostructure and tunable optical properties due to SPR

Surface plasmon resonance (SPR) is a resonance effect that occurs through the interaction of incident electromagnetic radiation with the conduction electrons (free electrons) of metal nanoparticles (NPs).[23,25,29,59] In an NP, a large fraction of atoms is the surface atoms and free surface electrons (*i.e*. conduction electrons) can collectively oscillate to produce the **surface plasmon (SP).** Due to quantum confinement (QC), the surface plasmon is localised and it is called the **localised surface plasmon (LSP).**[15,17,18,23,54] When a such metal nanoparticle is subjected to light, the oscillating electric vector of the electromagnetic field of the incident light can induce a collective oscillation of the free surface electrons with respect to the fixed position of the positively charged core. In other words, the incident light can cause shifting of the surface free electrons collectively to one side of the positively charged core to establish a dipole moment which oscillates (*i.e*. **dipolar oscillation** of the surface electrons). The **dipolar oscillation** occurs along the direction of the electric field of the incident light.

In SPR, the conduction electrons of surface atoms of the metal nanoparticles undergo a collective coherent or resonant oscillation when excited by the incident light at specific frequencies. In other words, when frequency of the incident electromagnetic radiation matches with the inherent frequency of surface plasmon, SPR occurs. Such metallic nanoparticles (NPs) showing the SPR excitation are called the plasmonic NPs (*e.g*. Au-, Ag- and Cu-NPs). The SPR causes both the strong absorption and scattering of light by the plasmonic metal nanoparticles (NPs). The SPR wavelength can be tuned from the UV–visible range to the near-infrared (NIR) regions by changing the size, shape, and composition of the plasmonic metal nanoparticles .[21,22] In this regard, the plasmonic NPs are comparable to QDs. The ratio of absorbing power and scattering power depends on the size, shape and composition of the plasmonic metal NPs. For the Au-NPs, the absorption is important for the smaller sized NPs (up to 30–40 nm) while the scattering becomes predominant for the larger NPs (>70–80 nm).[21,22] For the intermediate size range (about 60 nm), both the absorption and scattering are of comparable strength and such Au-NPs show the **dichoric behaviour**. The absorption and scattering power of Au-NPs due to SPR are much higher than those of similar sized nonplasmonic particles. Gold nanospheres (about 40 nm) show the absorption efficiency about 10^3 times higher than the conventional organic dyes showing the charge transfer (CT) bands and Au-nanospheres (about 80 nm) show the scattering efficiency about 10^5 times higher than the light emitted by the strong fluorescing dyes.[21,22] Such high scattering Au-NPs are more suitable for cellular imaging based on the light scattering microscopy while the high absorbing Au-NPs are more suitable for photothermal therapy[15] where the absorbed light energy is converted into heat that can destroy the cancer cells which are more heat sensitive than the normal cells.

The SPR properties of plasmonic nanoparticles like Au-NP, Ag-NP and Cu-NP have been extensively studied. The SPR absorption spectra depend on the size and morphology of the nanoparticles for a particular metal.[21,22] For gold, the optical properties of Au-nanospheres, Au-nanorods, Au-nanoshells (*i.e.* SiO_2@Au), hollow gold nanoparticles (HGNPs), etc have been studied extensively. The SPR wavelength can be more easily tuned upto the near infrared (NIR) range for the Au-nanorods, Au-nanoshells (*i.e.* SiO_2@Au), hollow gold nanoparticles (HGNPs) than for the Au-nanospheres for which the SPR wavelength can be tuned within the visible range only. The gold nanostructures having the SPR wavelength in the NIR range which represents the biologically transparent window are suitable for bioimaging. Because of the **tunable optical properties**, the plasmonic NPs are so important and they are the promising alternatives to the toxic metal based QDs in cellular imaging.

Besides the shape and size dependence of SPR wavelength, it shows the dependence on the refractive index of the surrounding environment. With the increase of refractive index of the surrounding medium around the Au-NP, the SPR wavelength experiences a red shift.[15] Thus the SPR wavelength of Au-NPs can be tuned also by surface coating with appropriate shells because the SPR wavelength depends on the refractive index of surrounding of the gold nanostructure. This property makes the Au-NPs **excellent**

molecular sensors and dielectric sensors. In fact, gold nanoparticles (GNPs) can be used in biological sensing because on binding of the biological analytes on the surface of GNPs, there is a significant shift in SPR wavelength.[15]

Due to the SPR absorbance, Au-NPs look from *ruby-red to violet to blue* depending on the size (red shift of the SPR λ_{max} with the size) and Ag-NPs are characterized by a *yellow colour*. **Faraday's red gold sol** (prepared in 1857 by M. Faraday through the reduction of $H[AuCl_4]$ by phosphorous in CS_2 solvent that probably acted as the stabilizer) is due to the SPR absorption by the smaller sized Au-NPs. Here it is worth mentioning that ***Faraday's red gold sol (preserved in the Royal Institution, London) was stable for more than one hundred years*** and it was unfortunately destroyed during the 2nd World War due to bombardment. Freshly prepared Cu-NP solution looks red.[60] The deep red colour of Cu-NP solution is due to ***the strong SPR absorption*** in the range from 400 nm (violet) to 620 nm (orange) (*i.e.* transparent to red light exceeding 630 nm wavelength).[17] Here it is worth mentioning that in contrast to Au- and Ag-NPs, *the surface* **plasmon resonance (SPR)** *of Cu-NPs does not strongly depend on the morphology of the Cu-NPs due to the strong influence of the 3d electrons*.[60] Relativistic effect may be the reason behind this observation. For the heavier congeners silver and gold, their higher *d*-orbital electrons are more destabilised due to the ***relativistic expansion of the higher 4d- and 5d-orbitals while for the lightest congener copper, the 3d orbitals are more strongly stabilized because the relativistic effect for copper is almost insignificant***.[13,14] In other words, the 4*d*- and 5*d*-electron clouds of the heavier congeners are more polarisable than the 3*d*-electron cloud of copper.

Like the photonic structural colour, the plasmonic structural colour can survive for a long period without any chance of photobleaching. On the contrary, the colouring with the chromophore based organic pigments becomes faded due to photobleaching on standing. This is why, this plasmonic nanotechnology of colouring has been attractive for colouring in ceramic industries.[7,11,26,31,35,44]

4.2 Plasmonic structural colouring in glass and ceramic products

In fact, since the ancient times, plasmonic structural colouring has been used by the glassmakers to achieve the vibrant colours by doping glass with the plasmonic metal nanoparticles to produce the beautiful objects. The science behind this technology was not known to them.[15,59] The classical examples of the application of this technology are: the **Celtic red enamels** (dated from 400 to 100 BC), **Roman Lycurgus Cup** (dated to the 4th Century AD) and **stained glass** developed in the late Bronze Age (1,200–1,000 BC), etc. The use of stained glass in the medieval ages (5th Century AD–15th Century AD) is now well established.

A trace amount of copper nanoparticles (Cu-NPs) is responsible for red colour of the ***Celtic red enamels***. Here it may be noted that the freshly prepared plasmonic Cu-NP solution (obtained through the reduction of dilute $CuSO_4$ solution by $NaBH_4$)[60] is wine red (fresh Cu-NPs). But on standing, it turns to yellow (oxidized Cu-NPs, Cu_2O, indicating the high reactivity of metallic Cu-NP) or dark green and blue (agglomerated Cu-NPs). It may be noted that to prevent the oxidation of Cu-NPs in manufacturing the red glasses, tin is added to the glass as a reducing agent (*cf.* Sn(II) compounds act as the reducing agent for copper). Iron is also a well-known reducing agent to reduce copper.

Besides the Cu-NPs, the Ag-NPs and Au-NPs were also used in the ancient Roman glasses for colouring. In fact, medieval cathedral red windows were stained with the Au-NPs. ***It is now established that both Cu-NPs and Au-NPs can act as the colouring agents in ruby-red glasses.*** The Cu-rich nanoparticles contribute to the purple colour in glass. Moreover, the deepness of colour depends on the variation of concentration of the plasmonic NPs. ***It may be noted that the photonic structural colour is found in nature but the plasmonic structural colour does not occur in nature***. In the ancient times, the glassmakers used the plasmonic nanotechnology for colouring but the science behind it was not known to them. Now it is established that the ***vibrant colours of these historic artefacts result from the SPR effect of metallic NPs***. This science has been revealed only in recent times.

4.3 Dichoric properties of gold and silver nanopartices and Lycurgus cup

The ***dichroic behaviour*** of **Roman Lycurgus Cup** (a Roman glass artefact) is historically famous and it is now preserved in British Museum (https://www.britishmuseum.org/).[8] This scientific serendipity has puzzled the scientists for centuries.[19,28] It shows a **red colour** when a white light source illuminates it from inside the cup but it shows a **green colour** when the light source illuminates from the outside (Figure 6). ***Thus the cup reflects the green colour but it transmits the red colour.*** A dichroic material exhibits two different colours when it is illuminated from the front side (*i.e.* reflection, more correctly back-scattering) and from the back side (*i.e.* transmission) (*cf.* **dichroism:** the property of exhibiting different colours by the reflected/back-scattered and transmitted lights; a colour change because of a change

Figure 6: Images of the dichoric Lycurgus cup, a Roman artefact kept in the British Museum. Green colour (left side) under reflected/scattered light (*i.e.* illumination from the front-side) and transmitted light (*i.e.* illumination from the in-side)'. Both images reproduced, with permission, from British Museum. Copyright Trustees of the British Museum, used under a CC BY-NC-SA 4.0 license.

in the illumination direction). This dichoric property of the cup is a result of the distinct transmission and scattering spectra produced by the plasmonic Au- and Ag-NPs of different shapes and sizes embedded into the cup.[16,27,28]

For the Au-NPs, it has been shown that the extent of Mie scattering responsible for the reflection colour increases with the increase of NP size. In fact, the relative contribution of scattering and absorption to the total extinction is highly size dependent. For the smaller Au-NPs (*ca.* 10–30 nm), the contribution of absorbance to the total extinction is the predominant one and the relative contribution of scattering increases with the increase of size of the Au-NPs, and the reverse is true for the larger Au-NPs (>80 nm).[21,22] Thus the medium sized (*ca.* 60 nm) Au-NPs possessing both the absorption and scattering efficiency comparable can show the dichoric behaviour (*i.e.* purple/violet absorption colour and brown back-scattering/reflection colour) while the smaller Au-NP (*ca.* 10–30 nm) solution for which the scattering effect is negligible is **nondichoric** (red coloured due to the predominant SPR absorption). Dichroic Au-NPs (ellipsoidal shaped, 50–60 nm) have been synthesised in a modified Turkevich method[16,27,28] by using a sub-stochiometric ratio of reducing agent (citrate to $H[AuCl_4]$ molar ratio ≈ 0.7) compared to a standard Turkevich method (citrate to $H[AuCl_4]$ molar ratio ≈ 10) that generates the citrate stabilized nondichoric Au-NPs of smaller size (≈20 nm). The ellipsoidal Au-NP (50–60 nm with a mean aspect ratio of 1.4) solution is dichoric and it shows a **brown colour in reflection due to back-scattering**, while the solution looks **violet coloured in transmission**[16,27,28] (Figure 4). Similarly, the smaller Ag-NPs look yellow due to the predominant SPR absorption but the relatively larger Ag-NPs (synthesised by the reduction of $AgNO_3$ by $Na[BH_4]$ and polyvinylpyrrolidone (PVP) as the stabilising agent) for which the Mie scattering effect is quite important show the dichoric behaviour (*i.e.* Ag-NP@PVP solution shows a **green colour in reflection due to Mie scattering**, while the solution looks **yellow coloured in transmission)** (Figure 7). The **Lycurgus Cup effect** has been reproduced in laboratory by placing a cuvette with a red nondichoric Au-NP (*ca.* 20 nm) solution behind the dichoric Ag-NP solution. This combination shows the *green colour in reflection* and *red colour in transmission* as in the **Lycurgus Cup.** It may be noted that mixing of the two Ag and Au nanoparticle solutions together will not produce the same dichroic effect because the two metal NPs are not strongly capped and may undergo metal exchange. Here it is worth mentioning that a 3D printed cup using a nanocomposite material having the ratio of Ag-NP@PVP (dichoric)/Au-NP@PVP (nondichoric) of 90:10 and a thickness of 600 µm shows the same dichroic effect of the Lycurgus cup under different illuminations.[27]

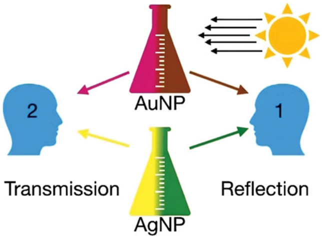

Figure 7: Dichroic behaviour of gold and silver nanoparticle (Au-NPs and Ag-NPs) solutions: two different colours depending on the positions of incident light and observer. Au-NP solution reflects the **brown light** and transmits the **violet light** while Ag-NP solution reflects the **green light** and transmits the **yellow light** (reproduced with permission from[16], courtesy of CC BY-NC-ND 4.0).

Lycurgus Cup (a masterpiece of Roman glass artefact, now kept in British Museum) depicts that the hot-tempered King Lycurgus (in Greek mythology) attempted to kill Ambrosia, a follower of the god Dionysus (god of vines and wine). Then she was transformed into a vine that wrapped around the king and restrained him. Eventually, she killed the king Lycurgus. Dionysus and two followers are also shown in the cup's glass taunting and irritating the king entangled in the vines of Ambrosia.

5 Conclusions

The chemistry behind the nanotechnology of colour generation by the quantum dots (QDs), nanostructured photonic crystals (PCs) and plasmonic metal nanoparticles has been discussed in this article (Figure 8). Due to quantum confinement (QC) in the zero dimensional semiconductor quantum dots (QDs), discrete energy levels are produced in both the CB and VB and bandgap (E_g) increases with the decrease of particle size. The dependence of bandgap (E_g) on the size is expressed by the Brus equation. Thus the QDs show the tunable and size dependent fluorescence emission properties with the characteristic features: broad absorption (excitation) band extending towards the UV region, a sharp and symmetrical fluoresence band, very high molar extinction coefficient and red shift of the emission peak with the increase of particle size. Their intense fluorescence colour can be varied from the UV–visible range to the near-infrared (NIR) range. QDs are important in colourful display and bioimgaing. Photonic crystals (PCs) are the nanostructured materials self assembled in a periodic manner to act as the diffraction gratings for Bragg's diffraction. The incident electromagnetic radiation is reflected from the different planes of PCs and the constructive interference among the reflected waves at some specific wavelengths and angle of incidence gives the strong reflection called ***iridescent colour*** which is material independent. The colour play in the opals and pearls, wings of peacocks and butterflies, in the necks of pigeons, variable colours in chameleons, etc. are due to the presence of photonic crystals (PCs). Plasmonic nanoparticles like Au-, Ag- and Cu-nanoparticles are characterized by the presence of free conduction electrons that can collectively oscillate to produce the surface plasmon that remains localized due to the quantum confinement effect to produce the localized surface plasmon (LSP). In SPR (surface plasmon resonance), these free conduction electrons of surface atoms of the metal nanoparticles undergo a collective coherent or resonant oscillation in the interaction with the oscillating electric vector of the electromagnetic field of the incident light at specific frequencies. The SPR causes both light absorption and scattering by the plasmonic NPs and the SPR wavelength can be tuned by varying the size, morphology, composition of the NPs and refractive index of the surroundings. Colouring of ceramics and glasses by the embedded plasmonic nanoparicles is well known.

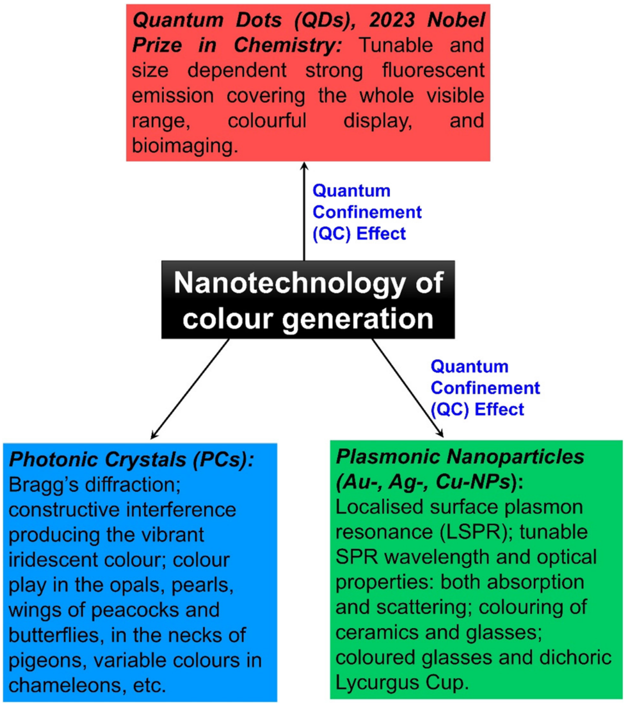

Figure 8: Illustration of the different mechanisms of the nanotechnology of colour.

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