

Why Are Bismuth Ions Interesting for Luminescent Solids Design?



Serhii Nedilko^{1*}, Victor Borisyuk¹, Vadim Sheludko³, Vitalii Chornii^{1,2}

¹Taras Shevchenko National University of Kyiv, Ukraine

²National University of Life and Environmental Sciences of Ukraine, Ukraine

³Oleksandr Dovzhenko Hlukhiv National Pedagogical University, Ukraine

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*Corresponding author: Serhii Nedilko, Taras Shevchenko National University of Kyiv, 01601 Kyiv, Ukraine

Abstract

The paper presents a brief overview of the luminescent properties of solids - crystals and glasses containing bismuth ions of different charges: Bi³⁺, Bi²⁺, and Bi⁺. The mentioned luminescent materials attract attention as promising for various high-tech applications. Among them are scintillator materials, phosphors, converters for solar cells and coatings for white LEDs. The review considers the factors that determine importance for applications characteristics of luminescent centers formed by bismuth ions in solids.

Keywords: Bismuth; Ions; Crystal; Glass; Luminescence; Phosphates; Sulphates; Materials; Solids; Niobates

Abbreviations: LCs: Luminescence Centers; UV: Ultraviolet Region; PLE: Photoluminescence Excitation; TRES: Triplet Relaxed Excited State; WLEDs: Wight Light-Emitting Diode

Introduction

Studies of the luminescence of solids containing bismuth ions have a long history, and their detailed systematic studies can be considered to have begun in the mid-1960s of the last century. Among the studied solids are "exotic" frozen solutions of alkali halide salts which developed by Prof. Mikle U. Bily at Optics Dept. of the Taras Shevchenko National University of Kyiv (Ukraine), various types of crystals (alkali halides; alkaline-earth oxides, sulphates and phosphates; tungstate's; silicates; borates; vanadate's; niobates, etc. [1-7], glasses (borate, phosphate, etc. [7-10]. Luminescent micro-nano-sized powders of bismuth-containing compounds are currently being studied as fillers in polymer flexible nanocomposites [11-13]. The last two decades have been characterized by a significant increase in interest in the development and large-scale study, both of experimental [14-19] and theoretical and computational methods [20,21], and references therein), of solid-state materials that exhibit luminescent properties due to luminescence centers (LCs) formed by bismuth ions.

The luminescence centers in solids are localized formations consisting of a center's core (ions of mercury-like, transition, and

rare earth elements, etc.) and a shell (atoms/ions/molecules/defects of the solid lattice) surrounding the core. The energy structure of the LCs core determines mainly luminescent characteristics of the LCs, but the characteristics of the LCs can be modified by the effect of the local environment and the symmetry of the latter. This feature allows for characterizing the LC as a luminescent probe of a structure of solids. Here, the mini review about properties of the luminescent solids where the bismuth ion is the core of the luminescence center.

Discussion

Luminescent bismuth containing materials exist in the form of bulk bodies - both crystalline [2-5] and amorphous/glass [8-10]; thin films - both mon- and multilayer [18,22]; and nanosized phosphors [23,24]. Among bismuth-containing materials there are those where bismuth ions are components of the lattice [3,6,7], and the dopants are triply charged ions of rare earth elements RE³⁺ (lanthanides). There are also such materials, on the contrary, RE ions are constituents of the lattice, and bismuth ions are dopants [2,4,5]. Both options are easily realized due to the same charges of Bi³⁺ and Ln³⁺ ions and the closeness of their effective ionic radii

(r). (In particular, as for Eu^{3+} : $r(\text{Eu}^{3+}) = 0.1066\text{nm}$ and $r(\text{Bi}^{3+}) = 0.117\text{nm}$, if they are coordinated by eight ions [24]. This fact (F1), along with other important useful properties, determines the continuous interest shown by researchers and technologists in the bismuth-containing luminescent materials [25-28]. The wide range of applications and the prospects for expanding this range are due to several other factors. One of them (F2) is the presence of broad and intense absorption bands in the ultraviolet region (UV) that allows for the intensive absorption of light and the effective excitation of the Bi³⁺ ions' luminescence.

Bi ions: background

The Bi atoms have the electronic configuration $(\text{Xe})4f^{14}5d^{10}6s^26p^3$, while Bi ions can be in charge states 0, +1, +2, +3, and +5 in a composition of the solids. Bi^{3+} ions have a $6s^2$ electronic configuration which is quite sensitive to influence of the nearest surrounding [29]. The ground state in this configuration is 1S_0 , and the excited states are 3P_0 , 3P_1 , 3P_2 , and 1P_1 in order of the energy increase (Figure 1a). According to the selection rules for electro-dipole $^1S_0 \rightarrow ^3P_0$ and $^1S_0 \rightarrow ^3P_2$ transitions are strictly prohibited. The dipole $^1S_0 \rightarrow ^1P_1$ transitions are only allowed. The $^1S_0 \rightarrow ^3P_1$ transitions are partly allowed due to the 1P_1 and 3P_1 states mixed by the spin-orbit interaction. In the solids, the forbidden $^1S_0 \rightarrow ^3P_2$ transitions also become partly allowed due

to electron - vibronic mixing. Thus, in the solids, the absorption (photoluminescence excitation) bands (PLE) caused by the electronic transitions from the ground 1S_0 level of a free Bi^{3+} ion to the excited 3P_1 , 3P_2 , and 1P_1 levels can be observed. Usually, the bands are labeled as A, B, and C (Figure 1a). Energies of A, B, and C transitions are near 9.4, 12, and 14.2eV, respectively, for free Bi^{3+} ions, and they are lowered by nearly 2 times under crystal force influence and spin-orbit interactions in the solids [29,30].

Luminescence of bismuth in crystals

The Bi-doped crystals due to the presence at ambient conditions of an intense Bi^{3+} - ions related luminescence are prospective materials for scintillators, X-ray screens, etc. [4, 21, 22, 31]. The co-doped Bi^{3+} and trivalent rare-earth ions (Ln^{3+} : Dy^{3+} , Er^{3+} , Yb^{3+} , Eu^{3+} , Sm^{3+} , Ho^{3+} , Nd^{3+}) materials were found to be potentially applicable as spectral converters for solar cells and wight light-emitting diodes (WLEDs) [31-38]. The Bi^{3+} ions possess the $6s^2$ electronic configuration, which is quite sensitive to the influence of the nearest surrounding. The Bi^{3+} ions luminescence occurs due to $^3P_1 \rightarrow ^1S_0$ radiative transition, which causes the spectral band of visible "blue" PL, and the peak positions of this band can lie between 425 and 570nm, that is determined by the interaction of Bi^{3+} ions with the environment, by the structure and symmetry of the nearest neighbor's location (Figure 1b).

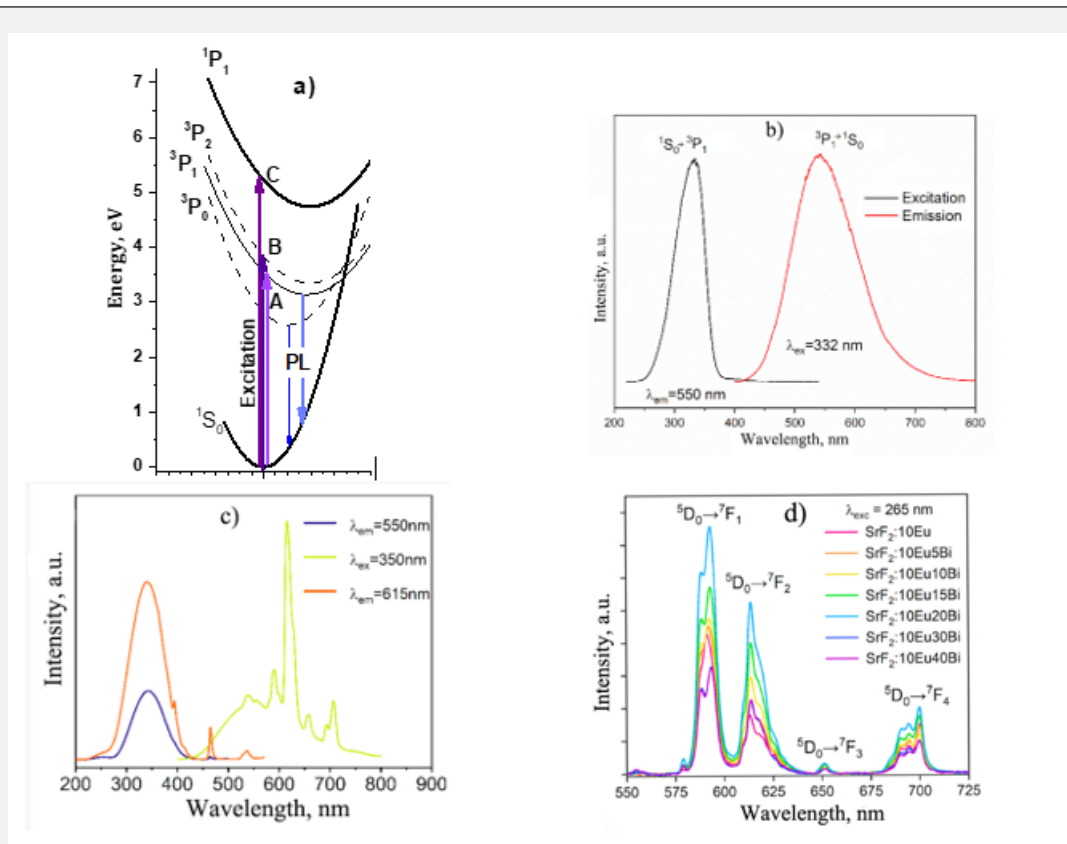


Figure 1: Energy levels scheme of the Bi^{3+} ions in solids (a); the PL and PL excitation spectra of the $\text{LuVO}_4:1\%\text{Bi}$ [17] (b), $\text{SrLa}_2\text{Sc}_2\text{O}_7:6\%\text{Bi}^{3+}, 5\%\text{Eu}^{3+}$ [18] (c), and $\text{SrF}_2: \text{Eu}, \text{Bi}$ phosphors [19] (d).

Only the A band clearly appears in the absorption /excitation spectra of many solids. The peaks positions of the A band and related band of the PL excitation strongly depend on the type of solid. So, as the data published show, the A - position can vary from 210nm (5.86eV) to 450nm (2.7eV) [4] (Figure 1b & 1c).

It had been shown many years ago [39-41], two types of Bi^{3+} - related emissions are revealed in the crystalline materials [1,4,41,42]. The PL of both types arises from the triplet relaxed excited state (TRES) of a luminescence center. Due to the spin-orbit interaction, the TRES is splitting and arranges two emitting levels. (Radiative transitions are indicated as the PL arrows on the simplified diagram (Figure 1a). The distance (D) between these levels depends on the energy of the spin-orbit interaction. The probability of radiative transitions from the top level is much higher than that from the metastable bottom level. Thus, the luminescence decay shows two various components, and the temperature behaviors of the decay times strongly depend on the D value [4,43].

The PL bands noted above differ by the Stokes shifts (S) and full widths at half maximum of the emission bands (FWHM), and by the temperature dependences of the luminescence decay time. The higher-energy emission band of Bi^{3+} - related centers, located usually in the ultraviolet (UV) spectral region (UV emission), is characterized by relatively small values of FWHM (0.14 - 0.38eV) and S (0.25 - 1.4eV). The decay time of the slow component of this PL is temperature-independent up to high temperatures (40-100K) indicating the large D values. The FWHM and S values of the lower-energy emission band (usually located in the visible (VIS) spectral range - VIS emission) are much larger: 0.5 - 1.0 and 1.2 - 2.6eV, respectively. The slow component decay time is temperature-independent in narrow temperature diapason 1-5K, and D (0.4 - 5meV) and S values (47 - 265meV) are 1-2 orders of magnitude smaller if compared to the UV emission (0.14 - 0.38eV) (0.25 - 1.4eV) [4].

There are a lot of compounds where both types of PL bands co-exist, and there are compounds where only the VIS emission was observed or, on the contrary, only UV band was manifested [4]. The indicated diversity of luminescent manifestations of LCs formed by Bi ions in various materials is due to the structure of the electron shells of bismuth atoms/ions, the type of electronic transitions between the states of free ions (mostly forbidden!), and the lifting of prohibitions on transitions for ions in the lattices of solids. This is the third factor (F3), which explains the Bi - materials usefulness and prospects.

That is, due to the presence of broad and intense absorption bands in the UV region, intense wide visible PL bands associated with Bi, and also due to an efficient excitation energy transfer

from Bi^{3+} to RE^{3+} ions (F3 factor) an intense visible emission occurs from RE^{3+} ions (Figure 1c & 1d) as well as the references [4,16,34-38, 44-47]. Luminescent manifestations of bismuth can be complicated by the presence of bismuth ions in other than 3^+ charge states, particularly those can be the Bi^{2+} , Bi^+ , and Bi^0 states. Thus, the PL and PLE transitions in Bi^{2+} and Bi^+ ions are shown in Figure 2a. Due to the p-type of the ground and lowest excited levels, which are involved in the radiative dissipation of the excitation, Bi^{2+} ion becomes sensitive to the surrounding crystal field. As a result, the band's peak positions of Bi^{2+} PL can be located at various peak positions in the range from 550 to 720nm [48-51]. More data about the PL properties of the Bi^+ ions and Bi^0 LCs in crystals can be taken in [48]. The formation of Bi ions of different charges and the possibility of controlling their composition through crystal-chemical procedures is another (F4) important and attractive feature of bismuth-containing crystals.

Luminescent bismuth containing glasses

Glasses are the mediums where the environment of bismuth ions can vary from ion to ion. In a glassy medium, such an ensemble is described by a set of energy levels and luminescent characteristics. This phenomenon is called optical inhomogeneous broadening of spectral lines, and it can lead to strong changes in the PL mechanisms and Bi - ions the PL characteristics compared to ones in crystals.

Today, bismuth-containing oxide glass has become an important technological material due to their structure and physical properties such as low melting temperatures, high densities, high refractive indices, high thermal expansion, low transformation temperature, excellent infrared transmission, and large attenuation coefficients for X-ray and gamma radiation. These glasses can be used for electromagnetic radiation shielding, as thermal and mechanical sensory, ultrafast optical switching, induced/laser generation of light, etc. [9,52-59].

Luminescent manifestations, like the described above for Bi - containing crystals, were observed for bismuth-containing glasses of different structure and composition [8-10]. The absorption and luminescence spectral features have been associated with electronic transitions in ions/atoms/groups such as Bi^{5+} , Bi^{3+} , Bi^{2+} , Bi^+ , Bi^0 , $\text{Bi}^{2-}/\text{Bi}_2^{2-}$, $\text{Bi}^{2-}/\text{Bi}_2$, Bi and their clusters [26-28]. Earlier, it was also emphasized that the formation of several bismuth ions of different charge states could be the result of reversible auto-thermal reduction of Bi^{3+} ions down to the Bi^0 state, which can occur during melting, or one of the transformations of intermediate valence states: $\text{Bi}^{3+} \leftrightarrow \text{Bi}^{2+} \leftrightarrow \text{Bi}^+ \leftrightarrow \text{Bi}^0$ [9,26-28]. The PL spectra related to the above noted ions are in the 550 - 1600nm region longer than the spectra of the Bi^{3+} (Figure 2), that was already described above regarding Bi containing crystals.

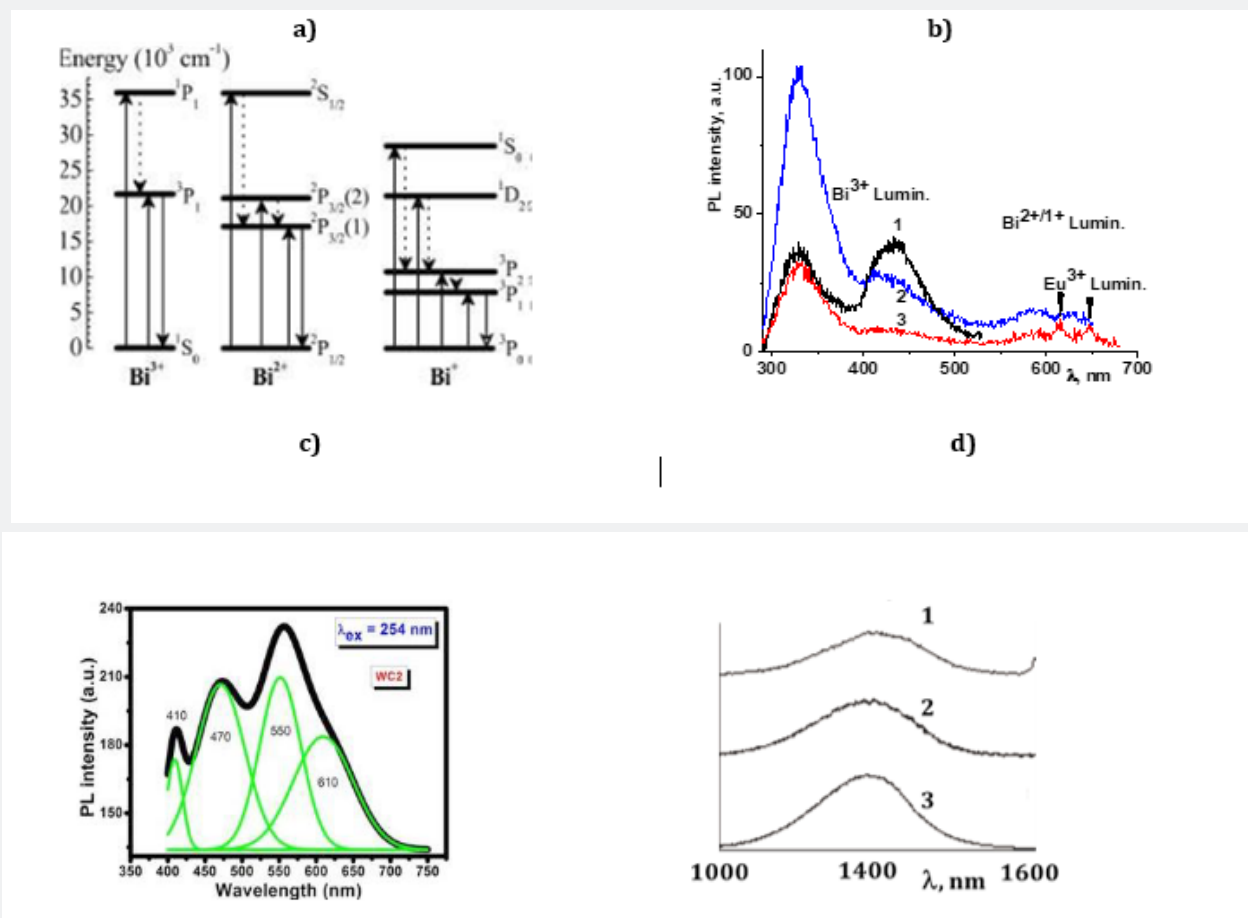


Figure 2: a) Energy levels scheme of the Bi^{3+} , Bi^{2+} , and Bi^{+} ions in glasses [55]; b) PL spectra of $\text{KBi}(\text{MoO}_4)_2$ phosphor (1), $31.79\text{P}_2\text{O}_5$ - 25.43MoO_3 - $37.09\text{K}_2\text{O}$ - $5.69\text{Bi}_2\text{O}_3$ glass (2), and the same glass doped with Eu^{3+} ions (3) excited at 275nm; c) PL spectra of 8WO_3 - $30\text{Bi}_2\text{O}_3$ - 2CeO_2 -(60) Bi_2O_3 glass excited at 254nm [59]; d) doped with Bi_2O_3 (1mol%) 33SrO - $67\text{B}_2\text{O}_3$ (1), 25SrO - $75\text{B}_2\text{O}_3$ (2), and 20SrO - $80\text{B}_2\text{O}_3$ glasses excited at 808nm [52].

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The relative contribution of these bands to the PL and PLE spectra depends on the ratio of the number of oxides forming and/or modifying the glass Figure 2b and [10,52,56]. Thus, the PL bands caused by the ²P_{3/2} → ²P_{1/2} transitions in Bi²⁺ ions show the peak positions near 560 and 690 nm (33SrO-67B₂O₃-1Bi₂O₃ glass) [52,55]; at 600 and 610nm (MO₃-B₂O₃-CeO₂-Bi₂O₃ (M= Mo or W) glasses [59]; (xMoO₃-30B₂O₃-(70-x)Bi₂O₃ glasses (x = 0, 2.5, 5, 7.5, and 10 in mol%) [9]; 640nm ((95-x)SiO₂·xSrO·5Al₂O₃·2Bi₂O₃ glasses (x = 30, 35, 40, 45, 50, in mol%) [56]; 660nm in the spectra of the Bi₂O₃-doped (1 mol %) MO-B2O3 (M=Ca, Sr, Ba) glasses. For the SrO-B2O3-Bi2O3 system, longer-wavelength luminescence lying in the near-IR region was also observed [53,55]. The emission of the 23B₂O₃-5ZnO-72Bi₂O₃-xCuO glasses in the range of 750-900nm (λ_{max} = 804nm) was attributed to ³P₂ → ³P₀ transitions in Bi⁺ ions (6s²6p² electronic configuration) [60].

Bismuth-containing glasses can exhibit the PL in the longer-wavelength IR region of 950-1600nm (Figure 2d). The nature of this emission is questionable, but it is undoubtedly also related to Bi centers. For example, it could be a luminescence caused by X₂²P_{3/2} → X₁²P_{1/2} radiation transitions in the BiO groups, as Bi₂O₃ can be partially converted into noted molecular species at high temperature [61].

Conclusion

Luminescence spectra of Biⁿ⁺ (n = 3, 2, and 1) - containing solids (crystals and glasses) include the overlapping emission bands related to radiative transitions in the Biⁿ ions related luminescence centers.

With a decrease in the Bi ions charge (Bi³⁺ → Bi²⁺ → Bi⁺), the absorption and luminescence bands of Bi-formed luminescence centers shift to the long-wavelength side of the light spectrum up to near the infra-red region.

- a) A diversity of the Biⁿ containing solids luminescence properties are related to:
- b) interchangeability of bismuth ions and rare earth ions in solids.

c) the presence of broad and intense absorption bands in the wide region of light (from ultraviolet to IR) that allows for the intensive absorption of light and the effective excitation of the Bi⁻ ions' luminescence.

d) neighborhood influence on the Bi⁻ ions charge states and structure of the Bi- related luminescence centers.

Multicolor luminescence from blue, green, and red to NIR region under different excitation wavelengths allows for the Bi – containing materials to be used as photonic materials of various applications, e.g., as wide-spectrum light sources.

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