The Role of Axions in the Formation of the Photoluminescence Spectrum in Dispersive Media

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Abstract: In the review, based on the analysis of the results published in the works of domestic and foreign researchers, a variant of an unconventional interpretation of the photoluminescence of dispersive media in the energy range of 0.5–3 eV is proposed. The interpretation meets the requirements of the energy conservation law for photons and axions participating in the photoluminescence process. The participation of axions in the process is consistent with Primakov's hypothesis. The role of nonradiative relaxation at the stage of axion decay is noted. The axion lifetimes are estimated for a number of dispersive media.

Keywords: photoluminescence; dispersive media; axion

1. Introduction

The review is devoted to photoluminescence (PL) and the axion problem [1–5]. The processes of interaction of light, usually laser monochromatic radiation with various media in the visible and near infrared regions of the spectrum (0.5–3 eV), are considered. The appearance as a result of the light exposure of a broadband PL at the output of the dispersive medium under study, in the author's opinion, can serve as evidence of the presence of axions in such processes. According to the existing theoretical concepts, the circumstance confirming the possibility of the presence of axions in the specified spectral region can be their decay, leading to the appearance of radiation at the output of the environment under study at frequencies that are absent at the entrance to the medium.

In this paper, the definition of an axion is based on Primakov's hypothesis [6,7], according to which the fusion of two primary photons in a strong electromagnetic field of an atomic nucleus can lead to the birth of a pseudoscalar particle (axion) and vice versa, a pseudoscalar particle (axion, neutral pion) can decay into two secondary photons—annihilation (forward and reverse) processes. The light beam, falling into the dispersion medium, loses laminarity due to the presence of atoms of the alloying element. When photons penetrate into the atom into the region of the nucleus, a meeting, a collision of photons becomes inevitable. The strength of the axion–photon interaction is determined by the energy characteristics of the outer and inner electron shells of an atom. If for the outer electron shells, the value of the binding energy of electrons is from several eV to several tens of eV, then for the inner shells of an atom, the order of this value is 102–104 eV. For the photon energy range that I am considering (0.5–3 eV), the interaction force turns out to be sufficient for the generation of axions.

For reference, we point out that in theoretical physics that there is no consensus on the nature of the axion. At this stage, all that matters to us is that the mass of the axion is small. This fact corresponds to the statement: "In the theory modified within the framework of the Grand Unification axion must be a particle of small mass" [4], apparently comparable in size to the small mass of a moving photon. Therefore, it becomes possible to extract information about the presence of axions in certain optical processes initiated by photons, which we will be guided by in the next part of the review.

This work was written by an experimenter. In our work, we do not limit ourselves to analyzing our own results. The availability of published experimental results obtained by many research groups in the visible and near infrared regions of the spectrum (0.5–3 eV) allows them to be compared and analyzed. In this paper, I discuss physical processes, mechanisms, and models that illustrate and confirm the position put forward by the author. After considering the materials included in the review, the readers will be offered a scheme according to which the axion lifetime estimates for a number of objects are estimated from published sources [1,7].

The traditional approach to the description of optical phenomena using wave representations [8] does not allow for the full interpretation of the results observed at the junction of optics and elementary particle physics. The involvement of methods used in the physics of the quantum world can help solve the problem. If we turn to photons (quanta), which successfully moved from particle physics to optics with the advent of lasers, then the connection between optics and the physics of the quantum world turns out to be real. This fact will help us get closer to finding out the nature of the axiom.

Here, it is appropriate to recall the generation of harmonics of optical radiation, parametric light generation. The authors of [9], the subtitle of which is "electromagnetic waves in nonlinear dispersive media (DM), argued that when using the quantum interpretation, the birth of harmonics and the appearance of combinational frequencies indicates the essential role of the processes of fusion or division of photons (light quanta), noting the need to take into account the laws of conservation of energy and momentum in nonlinear optical transformations. When the second harmonic is generated, two pump radiation photons merge in an elementary act. In a parametric light generator [10], the decay of a pump photon in an elementary act ensures the appearance of two photons at the output of a nonlinear medium: a signal and an idle one.

The purpose of this review is as follows: based on the analysis of experimental data published by foreign and Russian researchers, to present a scheme, a model reflecting the contribution of axions to the PL process. The analysis of the results obtained in the study of PL, according to the author [1,7,8], admits the possibility of the presence of axions at the stage of interaction of pumping radiation with atoms used for doping the medium. As a rule, the medium under study is a dispersive medium (glass or crystal) in which the pump radiation is transformed, leading to the appearance of PL quanta. The process of conversion of pumping radiation is associated with the birth of axions and their decay. The energy of the PL quanta, as a rule, is different from the energy of the pump radiation quanta.

In our review, we will deal with a DM. The continuous phase DM consists of glass, crystal structure, and liquid. The second component of DM distributed (suspended) in the volume of the continuous phase consists of atoms or nanoparticles of alloying material: atoms of holmium, bismuth or silicon. That is to say, this phase consists of electrons and nucleis of atoms of the alloying material.

From chemistry, it is necessary to also recall the complex environment [11]. The complex environment is a substance composed of complex particles (atoms or nanoparticles) capable of independent or connected existence in a liquid, amorphous (glass), or crystalline environment, acting as a matrix. The complex particle itself, in turn, can be heformed from other, simpler particles (nuclei, surrounded by electrons). In optics, it is natural to talk about a dispersive medium—DM, a medium that is associated with the phenomenon of light dispersion.

Returning to the problem we are interested in, we inform you that the review will consider PL in DM, that is, in glasses doped with bismuth and holmium atoms, in crystal samples doped with holmium, and in PL suspended in ethanol silicon nanoparticles.

2. The Bohr Frequency and Its Relation to the Lorentz Harmonic Oscillator Model

In this section, we will consider the conditions for the propagation of a light beam in DM. In particular, these are the reasons that change the laminar propagation of a light beam to a turbulent one. This circumstance can change the phase velocity of photon propagation

in the DM, which leads to the interaction of primary photons with each other. Multiphoton nonlinear optical processes are considered as an example of such interaction.

2.1. Niels Bohr's Postulates

When considering the atom model, Niels Bohr proposed the postulates of quantization. Following [12,13], we recall their contents. The first postulate is that there are a certain number of electron orbits in an atom, which they call stationary states (levels). Moreover, in each of these states, an electron can stay indefinitely without radiating at all. This position was postulated arbitrarily and obviously contradicted the classical electromagnetic theory. Second, he postulated that an electron could be knocked out by a blow from the lower stationary, normal state E_1 to an energetically higher E_2, which Bohr called excited. The transition between these states is characterized by the Bohr frequency—ν_{21} [14], the value of which is determined from Equation (1)

$$\nu_{21} = (E_2 - E_1)/h, \tag{1}$$

h is Planck's constant, and $(E_2 - E_1) = \Delta E$ is the energy of the electron transition between levels. Such relations can link many levels together in pairs, and the role of the lower state can be assigned not only to the lower level but also to the higher ones. The Equation (1) in this case should have the following form:

$$\nu_{mp} = (E_m - E_p)/h \tag{2}$$

i corresponds to the level located below, and j corresponds to the level, located above. Thus, $m = 1,2,3 \ldots$, and $p = i + 1,2,3 \ldots$

Bohr's ideas proved useful for explaining the structure of optical spectra and found application in practical spectroscopy.

The next part of the review is devoted to the consideration of the processes of interaction of a monochromatic radiation beam with resonant and almost resonant media, which link the frequency of exciting radiation at the entrance to the medium to the frequencies of the medium levels responsible for scattered radiation at the exit of the medium under study with Bohr's frequencies. This circumstance occurs, as a rule, in nonresonant conditions, when the detuning of the pump radiation frequency relative to the frequency of the electronic transition in the ν_{mn} atom does not exceed 10–20 cm^{-1}. When considering the PL process, in various environments with volumetric placement of multidimensional oscillators, and, consequently, having a large set of Bohr frequencies, the proposed approach turns out to be useful for practical use.

As a rule, on spectrograms, the line corresponding to the Bohr frequency ν_{ij} is not observed at the output of DM.

2.2. The Bohr Frequency and Its Relation to the Lorentz Harmonic Oscillator Model

The Bohr frequency is a characteristic of the medium that appeared with the birth of quantum mechanics. Note that an expression similar to Equations (1) and (2 bnvvm) has been used in classical physics for a long time to describe the characteristics of a harmonic oscillator. We have to figure out how the refractive index n of a medium consisting of identical and different-sized oscillators behaves depending on the frequency. To do this, we should refer to the works that used a model in which the oscillations of electrons in an atom near the equilibrium position are considered [15–19] and compare with the work in which the author deals with the Bohr frequency [14]. In the first case, the book "Fundamentals of Optics" by M. Born and E. Wolf can help us [20]. Following to her we note that "the phase velocity V and, consequently, the refractive index $n(\nu)$ cannot be constant in the volume of the entire medium under study." These values depend on the detuning value of the pump frequency n relative to the Bohr frequency ν_{ij}. E. Fermi pointed out that the refractive index $n(\nu)$ and the phase velocity V are not quantities that have a constant value in the medium

under study [14]. For the classical harmonic Lorentz oscillator, according to [16] we have the Equation (2):

$$\nu_{mp} = (E_m - E_p)/h,$$

ν_{mp} is the natural frequency of the electron oscillations near the stable equilibrium position, E_m is the value of the electron energy at the "m" level, and E_p is the value of the electron energy at the "p" level [16]. Planck's constant $h = 6.6252 \times 10^{-27}$ erg $\times$ s.

According to Fermi [14], a similar ratio has the form:

$$\nu_{mp} = (E^{(m)} - E^{(p)})/\hbar \tag{3}$$

ν_{mp} is the Bohr transition frequency, $E^{(m)}$ is the energy of an electron occupying level "m", and $E^{(p)}$ is the energy of an electron occupying level "p". In this case, $\hbar = h/2\pi = 1.0544 \times 10^{-27}$ erg $\times$ s.

It is essential for us that, regardless of the type of model, the refractive index $n(\nu)$ depends on the pumping frequency.

Note that when photons pass through the electron shell surrounding the nucleus, both the magnitude and the sign of the pump frequency detuning relative to the frequency of the transition under study have an effect on the magnitude of their velocity.

If, for a harmonic oscillator, the refractive index $n(\nu)$ is greater than 1 (the pumping frequency ν is less than the Bohr frequency ν_{mp}, then the photon velocity slows down. If the value of the refractive index $n(\nu)$ is greater than the Bohr frequency ν_{mp}, then photons are reflected toward the pump light beam, which assists in the addition of photons, leading to the birth of an axion.

The results of consideration of the question of the relationship between the frequencies of the medium, its refractive index, and the characteristics of the radiation scattered by the medium are considered in detail in [21–26] on the example of atomic potassium vapors. Below, we will see that the considered dependencies will bring us closer to solving the problem: finding out the nature of the PL, the axion, and its place in the physical picture of the world. The Bohr frequencies are a real characteristic of the medium under study. The numerical values of spectral lines included in reference books and atlases are simple coincide with the Bohr frequencies of interlevel transitions in atoms. This position is valid for linear to laser optics. For high-power laser radiation used in experiments of this kind, shifts of electronic levels in atoms may occur [24], which naturally affects the spectral characteristics of scattered radiation and PL radiation at the outlet of the medium. As a rule, the level shift is associated with multiphoton processes. For example, in atomic pairs of potassium, these are three-photon electron Raman scattering [22] and six-photon parametric scattering (SPR) [25,26].

The frequency of the three-photon electron Raman ν_3 in a two-level medium can be calculated in accordance with the law of conservation of energy:

$$\nu_3 = 2\nu - \nu_{21} \tag{4}$$

where the multiplier h is omitted, and the frequency ν_{21} corresponds to the tabular value of the frequency of the interlevel transition.

Equation (4) does not take the shift of the levels of the medium under study in the pumping field into account; however, it allows comparing experimental data and tabular values of wavelengths or frequencies of interlevel transitions.

The results of experiments related to the three-photon electron Raman process can be found in [22–26].The similar results were obtain also in work [27]. Theoretical estimates of the three-photon process in two-level media are made in the book by Pantel R. and Puthof G. [18]. For further consideration, it should be noted that in the elementary act of three-photon Raman scattering Equation (4), two pump radiation photons participate. This fact will help us in solving the problem of PL and axion.

2.3. Starting the Analysis of the PL Spectra in the DM

In this section, we need to find out what happens to the radiation at the output of the DM under study, in which the atoms of the element alloying the medium are "weighted", while not forgetting that the atoms are nuclei surrounded by electron shells. Namely, the electrons of the shells determine the behavior of photons in the medium, and the behavior of the refractive index $n(\nu)$ DM near the resonance transition region is determined by electrons. Resonant transition and harmonic oscillator? According to Niels Bohr, there is a correlation of complementarity between "the unambiguous application of stationary states and the mechanical analysis of intra-atomic motions . . . " [12,13].

Taking this into account, we combine the ideas about the interlevel transitions of electrons of doping DM atoms with the oscillations of different-frequency harmonic oscillators. Namely, the electrons of the atoms doping the medium, responsible for the optical properties of the medium, are an ensemble of identical or different-frequency classical oscillators. Above, we have partially discussed the consequences of this circumstance.

For the future, we must consider three cases concerning the processes of interaction of the pump radiation ν with DM, namely:

(a) $\nu = \nu_{21}$, $n(\nu) \cong 1$;
(b) $\nu > \nu_{21}$, $n(\nu) > 1$;
(c) $\nu < \nu_{21}$, $n(\nu) > 1$,

where $n(\nu)$ is the refractive index of the medium at the frequency of the pump radiation ν; ν_{21}—the Bohr frequency resonant transition.

According to the theory of dispersion for the case:

(a) when $\nu = \nu_{21}$, the refractive index $n(\nu)$ of the medium is close to unity [15–18]. The reflection coefficient of such a medium increases as the excitation frequency ν approaches the Bohr frequency ν_{21}, reaching a maximum value at $\nu = \nu_{21}$ [16]. A qualitative illustration of this circumstance is Wood's experience in observing resonant radiation in the case of atomic sodium vapors placed in a cuvette illuminated by a sodium lamp. This experience practically illustrates the case of exposure to a two-level medium of photons whose frequency ν is equal to the Bohr frequency ($\nu = \nu_{21}$).

In fact, the scheme of forced (induced) photon emission proposed by A. Einstein in 1916 was implemented. Indeed, in a resonant two-level medium, in the elementary act of photon absorption, an electron leaves the ground level to the excited level.

By the next photon of the beam, electron can be knocked out from the excited level, i.e., we have:

$$h\nu_{21} + h\nu = 2h\nu \tag{5}$$

Here, $\nu = \nu_{21}$, where $\nu_{21} = (E_2 - E_1)/h$ is the transition frequency of the investigated duplex environment; the value of $h\nu_{21}$—energy of an electron in the excited level ($h\nu_{12} = h\nu_{21}$).

The left part of the Equation (5) open mechanism of enhancement of light emission in the inverted environment by doubling the number of photons, right side of Equation (5) in each elementary act of forced (induced) radiation. This process is implemented in tasks related to the amplification and generation of monochromatic radiation [20,28]. It should be noted that even at the dawn of the development of laser physics, the model of the classical harmonic Lorentz oscillator that we have attracted has already been used to consider the laser generation process [29].

In case (b), when $\nu > \nu_{21}$, the feature limiting the propagation of photons is due to the fact that, according to the theory of dispersion, the refractive index $n(\nu) < 1$. The propagation of photons of monochromatic radiation in this region of the spectrum, generally speaking, is impossible, because otherwise their velocity $V = c/n(\nu)$ would exceed the speed of light c, which contradicts existing concepts. If a high-power laser is used in the experiment, then due to three-photon electron Raman scattering [22] after leveling the populations of the levels of the transition under study, we will get $n(\nu) \cong 1$, which will allow part of the pumping to pass through such a medium. If the intensity distribution across the cross-

section of a powerful beam is Gaussian (single-mode laser), then we have self-focusing at the output of such a medium [21].

Finally, consider case (c) $\nu < \nu 21$, where $\nu 21$ is the resonant transition frequency. In this region of the spectrum, the refractive index is n(ν) > 1. According to [15–19], the refractive index n(ν) increases with decreasing detuning in the low-frequency spectral region with respect to the resonance frequency. This case was considered in [22]. Below is a summary of it.

If the condition $\nu \neq \nu 21$ is fulfilled in an elementary act, the energy of two pump photons ν should add up, which forms a kind of "virtual" level in the medium $(\nu + \nu) = 2\,\nu$, the breakdown of which is accompanied by the emission of one light quantum at a new frequency. PL is associated with this process. The fate of the second quantum is nonradiative relaxation (electron leaves "virtual" level for transition on one of real levels of doping atom—heating of the medium) [19].

When using high-power laser radiation with a frequency close to the transition frequency, the populations of the ground and excited levels of the atomic medium can be equalized due to three-photon electron Raman. In this case, according to Equation (4) radiation at the frequency ν_3 is recorded on the spectrograms [22,27].

When summing up the results under point (c) we note, that the process of three-photon electron Raman process is accompanied by a decrease of the refractive index of the medium $n(\nu) \rightarrow 1$ at the frequency of exciting radiation, ν.

If the pumping of the medium under study is carried out by a single-mode beam, in the cross-section of which the energy distribution has a Gaussian structure, then the consequence of this is a sagging refractive index on the beam axis. The consequence of this is a deviation from the axis of the particle beam of the pump radiation, which leads to the appearance of a conical structure of the beam at the outlet of the medium [21,22].

With an increase in the power of the exciting radiation, the process of three-photon electron Raman process is supplemented by a six-photon parametric process [25,26] in this case, and a second cone structure can be registered at the outlet of the cuvette with potassium vapor. The conditions of their appearance are discussed in detail. The first reason is the propagation of "superluminal" photons [23]. Naturally, the speed of propagation of these photons does not exceed the speed of light. The second reason is the propagation in a medium with superluminal velocity along the beam axis of nonlinear polarization associated with the six-photon parametric scattering [26].

We have shown that this situation can be explained by the dependence of the refractive index of the medium on the intensity of the pump radiation and the Gaussian intensity distribution over the cross-section of a single-mode beam. The propagation of photons in a medium with a phase velocity $V = c/n(\nu)$ makes them "superluminal" [30]. The results of experiments in atomic vapors of alkali metals in the frequency range of the main doublet can serve as a visual proof of the processes considered.

The propagation of radiation in a two-level medium occurs according to the law of conservation of energy. This applies equally to Raman and parametric multiphoton processes. The same correspondence takes place if we have an environment with a set of different frequency oscillators, which corresponds to DS. In comparison with the considered model of a two-level medium, in this case, due to a significant increase in the number of Bohr frequencies, the spectra of the radiation scattered by the medium become more complicated (broadening, overlapping). Nevertheless, the decoding of spectrograms turns out to be feasible within the framework of the proposed model.

The above information is necessary so that the reader can analyze the nonstandard.

3. Photoluminescence (PL)

In order to expand our understanding of PL, let us first dwell on the definition of this process. PL is luminescence excited by optical radiation [31]. Remaining within the framework of this definition, it is necessary to consider three options: (1) resonant radiation, (2) PL, corresponding to the Stokes rule, and (3) anti-Stokes PL.

The energy conservation law for PL [31,32] can be written in a form convenient for further work:

$$hv_{pl} = 2hv - hv_{ij}, \tag{6}$$

where hv_{pl}—the energy quanta (photons) PL, hv—the energy of the pumping quanta (photons)—the light radiation used to excite the PL, $2hv$—the energy of "virtual" level, v—the frequency of pumping radiation, and hv_{ij}—the energy of electron, expended for nonradiative relaxation.

The index i, j correspond to a set of levels involved in the relaxation process; index i—real level, index j—"virtual" level.

According to (6), each of these transitions corresponds to a component in the PL spectrum at the outlet of the medium. The value of v_{ij} is a combination of a large number of transition frequencies associated with nonradiative relaxation and transfer of thermal energy to the medium. Since the PL spectrum in DM is usually broadened, it is natural to assume that the PL process is accompanied by a multitude of electronic interlevel transitions responsible for nonradiative relaxation in the atoms of the element used for doping DM. Naturally, the law of conservation of energy is fulfilled for each frequency component of the broadened PL spectrum.

hv_{pl}—the quantum energy of the PL radiation,

v_{pl}—the frequencies filling the broadened spectrum of the PL.

The index "pl" corresponds to the whole set of frequencies involved in PL. The same number of frequencies of nonradiative transitions correspond to the index "ij".

From the Equation (6), it follows that the value of the pump radiation frequency is the arithmetic mean between each pair of frequencies v_{pl} and v_{ij}.

Resonant radiation is the simplest case of PL. In this case we have an implementation of the Einstein scheme: (1) absorption of a light quantum, (2) spontaneous emission of a quantum, and (3) stimulated radiation [17,18]. The experiments of the American optician R. Wood, who studied the resonant interaction of light radiation in a cuvette with atomic sodium vapor in the frequency range of the main doublet, were mentioned above. The process of absorption by sodium atoms of quanta falling on the window of a cuvette with atomic vapors is accompanied by the processes of spontaneous and forced resonant radiation.

Note that, in contrast to the resonant case, the PL spectrum at the DM output is, as a rule, broadened. Most often, it is broadened relative to the pumping frequency in the Stokes region of the spectrum. This fact is considered fundamental for the definition of PL. We will return to this issue below, since the processes of PL have not yet, in fact, been fully investigated. This was noted by Paul R.V. [17], referring to the experiments of Vavilov S.I. [33]. Finally, the well-known rule of mirror symmetry between the PL spectrum and the absorption spectrum is uniquely associated with the law of conservation of energy [28]. Indeed, in compliance with Equation (6) from the law of conservation of energy frequencies involved in the PL process, for the Stokes region of the PL spectrum, we have mirror symmetry:

$$v_{ij} - v = v - v_{pl} \tag{7}$$

here: $v > v_{pl}$.

A similar ratio can be written for the anti-Stokes region of the PL spectrum:

$$v_{pl} - v = v - v_{ij} \tag{8}$$

here $v < v_{pl}$.

The Equations (7) and (8) helped to author to understand the PL spectra, obtained by him [34–36], or borrowed from the materials of various authors, in which the results on PL are presented, and to which we will return below. Unfortunately, the accuracy of the frequency estimation at the stage of analysis of the PL spectrograms, published in the journals in the DM could not be high enough. However, this did not prevent the author from establishing coincidences between the tabular values of wavelengths or frequencies

responsible for nonradiative relaxation and the values of frequencies or wavelengths calculated from the experimental results.

If a frequency-nontunable laser is used as a pump, then the coincidence of the "virtual" level, whose energy is 2hν and the real level in the alloying atom is unlikely. That is why we do not have an exact match in the case of PL. In the next section of the paper, the case of an exact match is considered. The section is devoted to studies of PL and laser generation (LG) in holmium-doped media.

The next step is to discuss, within the framework of the above model, a number of experimental results published by domestic and foreign researchers and the consequences arising from the proposed model of PL.

4. On the Results of Studies of PL and LG in Holmium-Doped Media

We will focus on the studies of PL in DM performed in different laboratories. The purpose is to test the model, proposed by the author on the material of publications that contain information about the spectra of PL or laser generation (LG) in media activated (doped) with holmium. The author is aware that a small part of the publications on this topic has been considered. Nevertheless, the information extracted from the considered works confirms the position of the considered nontraditional model of PL.

The cases of using quartz glass fiber optical fibers [37,38], crystal samples, and chips [34,39–41] as DM are considered. Information about the preliminary review of the results of these works can be found in [42].

Both PL and LG processes starting at frequencies corresponding to the maximum peaks and humps on graphs reflecting the spectral dependence of intensity (power) will be considered PL at the DM output depends on the wavelength of the pump radiation. The transition of the overhead line to the LG mode in fiber light guides is carried out due to their length by retaining pumping and fluorescence radiation in the light guide. For chips, the LG mode is carried out using powerful pumping [34]. The use of holmium as an alloying additive is due to the search for media whose radiation is safe enough for vision, since the wavelengths of PL and LG radiation are in the IR region of the spectrum (~2 μm). In this area of the spectrum, it is convenient to conduct remote sensing, the development of differential absorption radars.

The reader should pay attention to the fact that to obtain PL in the infrared region of the spectrum (~2 μm), different sources whose wavelengths and radiation power differ significantly from different authors can be used for pumping.

The task of the analysis will be to determine the specific interlevel electronic transitions responsible for PL and LG in the specified spectral region (~2 μm). The use of holmium as an alloying additive is due to the search for media whose radiation is safe enough for vision, since the wavelengths of PL and LG radiation are in the IR region of the spectrum (~2 μm). Let us focus on the experiments performed using fiber light guides. Let us consider the results presented in the works of A.S. Kurkov and his collaborators [37,38]. According to these results, PL was obtained in the infrared region of the spectrum (~2 μm), and at wavelengths of 2.02 μm, 2.05 μm, 2.07 μm, 2.1 μm, 2.13 μm, 2.15 μm—LG. The experiments used a pump laser with a wavelength of 1125 nm, as well as 1147.35 nm. It is interesting to determine the wavelength of the electronic transition, which is associated with the heating of the glass fiber due to nonradiative relaxation. In particular, the transition responsible for LG in this region of the spectrum (~2 μm) corresponds, according to tabular data, to the wavelength value 755.09 nm [43]. This is the transition ^{5}I $(9 - 5^5/_2) - {}^4$I^0 $(^{15}/_2 - {}^9/_2)$ [44].

This result is obtained as follows. Previously, it was possible to obtain the value of the frequency ν_{pl}.

To calculate the frequency of the transition responsible for this process, it is necessary to use the following equation from the law of conservation of energy:

$$\nu_{pl} - \nu = \nu - \nu_{ij},$$

where:

ν is the frequency source of pump;

ν_{pl} is the frequency of the maximum peak on the spectrogam PL.

ν_{ij} is the frequency electronic transition between electronic levels (in case LG, for the holmium atom). Recall that the frequency ν_{ij} corresponds to transitions between any pair of electronic levels of opposite parity.

A slight (0.028 eV) discrepancy between the tabular data and the calculated value allows us to conclude that the results of the work [37,38] correspond to the PL model proposed by the author. Let us consider examples of obtaining radiation in the region (~2 μm) in crystal structures doped with holmium. In [41], when using a crystal made of yttrium–aluminum oxide (*YAlO₃*, *YAP*), radiation in the region (~2 μm) was obtained by pumping, the wavelength of which is 791 nm.

In [34], LG was obtained at a wavelength of 2050.5 nm (~2 μm) in a laser on a *Tm,Ho:YLF* microchip, when using a diode laser with a wavelength of 785 nm as a pump. In this case, the difference between the energy of the interlevel transition calculated according to Equation (7) corresponding to the frequency ν_{lm}, and, in fact, responsible for the PL, and the energy for the table value of the wavelength 4939.01 Å [43] does not exceed 0.043 eV. Thus, in the case of glass fiber for the 755.09 nm pumping wavelength used in the experiment, the transition, $^5 I\,(9 - 5^5/_2) - {}^4 I\,^0(^{15}/_2 - {}^9/_2)$, is responsible for the LG mode, corresponding to the frequency of 13243.35 cm^{-1}.

In the crystal structures, using the pumping 493,901 nm LG is responsible for transition $^5 I\,(9 - 5^5/_2) - {}^4 I\,^0(^{15}/_2 - {}^9/_2)$; its frequency—20246.97 cm^{-1}.

In [41], generation at a wavelength of 2044 nm was obtained using a holmium-doped *Tm, Ho:YALO₃* (*YAP*) crystal, where a laser diode was used for pumping, the radiation wavelength of which is 794.8 nm. Numerical calculation shows in this case that the difference between the calculated value of the energy of the interlevel transition responsible for the PL and the tabular value does not exceed 0.027 eV.

In [34], the case of obtaining LG at a wavelength of 2050.5 nm in a laser on a *Tm,Ho:YLF* microchip when using a diode laser with a wavelength of 785 nm as a pumping source is considered. The calculation shows that in this case, the difference between the calculated value of the energy of the interlevel transition corresponding to the frequency ν_{pl} and, in fact, responsible for photoluminescence, and the energy of the table value of the wavelength 4939.01 Å (see [43]) does not exceed 0.043 eV.

Thus, in the case of glass fiber [37,38], for the 755.09 nm pumping used in the experiment, the transition $^5 I\,(9 - 5^5/_2) - {}^4 I\,^0(^{15}/_2 - {}^9/_2)$, corresponds to the LG mode, corresponding to the frequency of 13243.35 cm^{-1}. In crystal structures, when pumping 493.901 nm, the transition $^5 I\,(9 - 5^5/_2) - {}^4 I\,^0(^{15}/_2 - {}^9/_2)$ is responsible for LG, its frequency is 20,246.97 cm^{-1}. Some results of the section are presented in [42].

5. Analysis of the Results, Observed in Bismuth-Doped Media

Are the results and conclusions of the previous section random? To make sure that the PL model we are considering is viable, let us consider the papers that present the results of the study of PL and LG in media doped with atomic bismuth. It is either fiberglass or monolithic glass. In fiberglass, as a rule, we are talking about PL and LG; in glass samples, we are talking about PL.

Table 1 presents information about PL and LG in the visible and IR spectral ranges. The criterion confirming the validity of our assumptions can be proximity (or even coincidence, which is not necessary) calculated transition frequencies associated with nonradiative relaxation. Naturally, the results of calculations of the frequencies of inter-level transitions are compared with the values of the frequencies presented in the reference literature, which has generally recognized fame and reliability [43,44].

Table 1. Juxtaposition tabular value of the transition wavelength with the result of calculating the wavelength of the transition.

1	2	3	4	5
Link number in the list literatures	Wavelength corresponding to the maximal valued ordinal curve of PL (LG): λ_{pf} (nm)	Wavelength of excitation source PL: $-\lambda$ (nm)	The result of calculating the wavelength of the transition: $-\lambda_{lm}$ (nm)	Tabular value of the transition wavelength: $-\lambda_t$ (nm)
			which is associated with non-radiative relaxation.	
[45]	~1130 nm	1058 nm	994.6 nm	982.8 nm
[46,47]	~720 nm	514 nm	399.6 nm	388.6 nm
[48,49]	~750 nm	500 nm	374.99 nm	359.6 nm
[48,49]	~1140 nm	500 nm	320.2 nm	323.9 nm
[48,50]	~1300 nm	800 nm	577.7 nm	574.2 nm
[48,51]	~1315 nm	808 nm	583.1 nm	574.2 nm
[48,52,53]	~1310 nm	808 nm	584.1 nm	574.2 nm
[48,54]	~1150 nm	980 nm	853.8 nm	854.4 nm
[48,55]	~1210 nm	405 nm	243.2 nm	243.3 nm
[56]	~1260 nm	798 nm	584 nm	574 nm
[56]	~1153.5 nm	502 nm	314.9 nm	306 nm
[56]	~1153,5 nm	525 nm	339.8 nm	339.7 nm
[56]	~1085.4 nm	680 nm	472 nm	472.2 nm
[56]	~1171.6 nm	738 nm	540 nm	555.2 nm
[56]	~1260 nm	798 nm	584 nm	527.4 nm

The works in which pumping was used in the 405–1230 nm spectral regions and the PL spectra were recorded in the 720–1650 nm range are considered. Bismuth atoms are embedded in a homogeneous amorphous isotropic medium (example: quartz, aluminosilicate glass), the temperature of which depends on the environment. For reference: bismuth has an ionization potential of 7.3 eV, which corresponds to 58,765 cm^{-1}.

The pumping radiation that initiates PL in a medium, containing bismuth atoms, due to the addition of two quanta of light in the elementary act, transfers electrons to "virtual" levels that occur near the levels of $^2D_{3/2}$, $^2D_{5/2}$, $^4P_{5/2}$, etc. These levels of bismuth correspond to the energy region of 0.5–3 eV.

The addition of two pump radiation quanta in the atomic nucleus field, according to Primakov [6], may indicate the birth of an axion. The condition for its appearance is the high intensity of intra-atomic fields. The "virtual" levels cannot be long-lived. The born axion decays into two new quanta, one of which leaves the medium—a contribution to the PL, and the second gives its energy to heating the medium due to nonradiative relaxation of electrons. Here, we should recall the direct and reverse effects of Primakov. Theorists are familiar with Feynman diagrams that illustrate the possibility of the existence of such processes.

In general, in bismuth-doped glass fiber, the structure of the PL spectrum can be quite complex, as evidenced by the published results [45–55]. Nevertheless, it is possible to distinguish frequencies that correspond to the maximum values of the spectral curve of the PL. The exact numerical value of the frequency corresponding to the top of the hump of the spectral curve PL is quite difficult to obtain from illustrations in journal publications. For this reason, in Table 1, depending on the pumping frequency, the discrepancy between the exact tabular value (column No. 5) of the transition frequency responsible for nonradiative relaxation and the one calculated on the basis of publications (column No. 4) cannot be ideal. However, the results of the calculation and the tabular values for the viewed publications are quite close to each other.

Column 1 of Table 1 contains a reference to the source. Column 2 indicates either the wavelength of the line on which the LG is obtained or the wavelength corresponding to the hump on the PL spectrogram. The wavelength of the excitation source PL is given in column 3. The calculated wavelength corresponding to the position of the hump having the maximum power value on the PL spectrogram (column 4). Tabular values of wavelengths of atomic bismuth lines with which radiative relaxation is associated are presented in column 5. The results from [45–55] were obtained in fiberglass, and only in the work [56] PL was investigated in a monoblock.

Before proceeding to the axion problem, let us briefly consider two more elements of the periodic system of Mendeleev, used for doping media used in optics and quantum electronics and related to PL.

First of all, I will focus on neodymium [57]. A new generation has come to replace the neodymium laser with lamp-pumped rods cooled by running water, in which laser LEDs are used to pump rods doped with neodymium. According to the author, the choice of the operating frequency of a narrow-band neodymium laser pump source corresponds to the PL model considered in the review.

A few words about silicon, the main material of electronics: The review [31,34,35] discussed the problem of mirror symmetry of PL and absorption of multi-dimensional silicon nanoparticles suspended in ethanol. The contribution of the anti-Stokes component in the PL spectrum is noted. The PL excitation of silicon nanoparticles suspended in ethanol was carried out using an argon laser $\lambda = 488$ nm.

The author [59], using lasers with different wavelengths (365 nm, 456 nm, 532 nm, 660 nm), showed that the peaks of the PL spectra (660 nm, 690 nm, 720 nm, 820 nm) of silicon nanoparticles are shifted, new sections of the spectrum are filled.

The relation (8) is valid in the case of LG. In the case of broadband PL, indices "*i,j*" correspond to electron transitions between the "virtual" level, whose energy is *2hν*, and the set of electronic levels of the doping atom. The frequencies of these transitions are not reflected in the tables and depend on the radiation frequency of the pump source. If the value of the pump radiation frequency ν is greater than the frequencies "ν_{ij}" of these transitions, then the conditions for anti-Stokes PL are met.

6. Axions in the Optical Range of the Spectrum and Their Lifetime

We have constantly noted above that when using monochromatic radiation to pump DM, the PL spectrum is, as a rule, broadened. The width of the PL spectrum is undoubtedly related to transitions in the atom of the alloying element. Among the reasons for the broadening of the PL spectrum are: the finite width of the electronic levels involved in the elementary act of excitation of the atoms of the element used for doping the medium, shifts of these levels in the pumping field [24], temperature conditions of the experiment.

The virtual level, whose energy is determined by the sum of the energies of the two pumping quanta, as a rule, does not coincide with the levels of the atom. According to Primakov [6], a meeting in an elementary act of two photons in the field of an atomic nucleus can lead to the birth of an axion. The time of its life is extremely limited. The instability of the virtual level due to the interaction with the levels of the atom of the alloying element in the field of the atomic nucleus leads to the decay of the axion into two quanta.

The energy of one of them is spent on heating the medium due to non-radiative relaxation. Unfortunately, the issue of heating, heat transfer of the DM is little discussed. But when dye lasers were being developed, the issue of heat dissipation was the main one. The same can be said about neodymium and other solid-state lasers [57].

As follows from the experiment, the main contribution to the width of the PL spectrum is due to the fact that the number of allowed transitions of electrons from the virtual level corresponds to the set of levels in the atomic shell. A lot of non-radiative electron transitions are the reason for heating the luminescent medium.

The second quantum of the decayed axion, according to the ratio (6), leaves the DM at a slow speed, which depends on the structure of the energy spectrum of the atoms of the alloying element, its energy, i.e., frequency. In addition, it is necessary to remember about the multitude of different-frequency harmonic oscillators (electrons) encountered on his way, and the law of dispersion for each of them. At the output of the DM, we get, as a rule, an expanded spectrum of PL. For example, when silicon nanoparticles placed in ethanol [34,58,59] are excited by argon laser radiation, a PL spectrum with a width from 464 nm to 670 nm is obtained.

What can the width of the PL spectrum give us and what information can we extract from it?

In fact, due to the quantum-mechanical uncertainty of the position of the energy levels of the atom, including the virtual one, we have the right to believe that the width of the PL spectrum recorded by the photodetector corresponds to the natural width [60]. If τ is the average lifetime of the axion at the virtual level, ΔW is the energy of this level, then for this situation there is a known uncertainty relation.

$$\Delta W \tau \approx h/2\pi \qquad (9)$$

According to [61], the uncertainty ratio due to the smallness of h is significant only for microsystems, which we are dealing with. Let us use this relation to determine the lifetime of the axion.

The contour of a wide spectrum of PL $I(v)$, reflecting the dependence of the power of the PL on the frequency, usually has a maximum (hump) and falling wings. For the width of the spectral line ΔW, a difference of frequencies is taken, which corresponds to a halving of its peak power $I(v)_{max}/2$. Table 2 shows several examples illustrating the relationship between the width of the spectral band of the PL radiation and the lifetime of the axion. To estimate the lifetime of the axion, the ratio was used: $1 \text{ cm}^{-1} \approx 2.99793 \times 10^{10} \text{ s}^{-1}$. The lifetime of the axion is significantly shorter than the lifetime of the excited energy levels, which, according to reference data [62], corresponds to a value of 10^{-8} s.

Table 2. Axion lifetime for Alloying Materials the Test Sample.

References	Alloying Material; in Parentheses— Environment of the Test Sample	The Wavelength of the Pump Radiation, nm	Frequency Corresponding to the Maximum Value of Intensity PL, cm^{-1}	Width of the PL Spectrum at Half-Length, ΔW, cm^{-1}	Axion Lifetime τ, s
[35,36]	Silicon (ethanol)	488	~5480	~1160	~3.48 × 10−13
[63]	Bismuth-Bi (glasscorderite))	514	~8547	~420 (~700) ~1120	2.93 × 10−13
[64]	Bismuth-Bi (aluminosil-rolled glass T = 77 K	1075	~8880	~1213	3.64 × 10−13
[65]	Bismuth-Bi (phosphorus-silicate glass)	1240	~7463	~340	~1.02 × 10−13
[66]	Bismuth-Bi T = 1.4 K	375	~6803	~340	1.02 × 10−13

7. Summing Up

Analysis of experimental results shows that the definition of PL in DM needs to be supplemented and clarified. We have previously settled on the definition according to which PL is a glow generated by optical radiation. By definition, Vavilov S.I. luminescence is an excess of radiation over temperature, provided that the excess radiation has a finite duration exceeding the period of light oscillations (10^{-10} s). Stokes's law states that PL light has a longer wavelength compared to the light used for excitation. According to Lommel,

the PL spectrum as a whole and its maximum are always shifted in comparison with the excitation spectrum and its maximum towards long waves.

For a long time, it was believed that the substance has a completely defined luminescence spectrum, which is not sensitive to changes in the wavelength of exciting light. For DM, this rule is violated. Experiments in silicon, holmium, and bismuth doped media have not confirmed this position. On the other hand, the validity of the rule of mirror symmetry of absorption and PL spectra established by Levshin V.L. is confirmed, which follows from the unconventional definition of PL, considered in the work.

Finally, the main thing: the laminar luminous flux of the pump radiation, when propagated in a dispersing medium, becomes turbulent, which allows photons to collide with each other, forming a virtual energy level in the atom shell. This circumstance ensures the appearance of axions with a short lifetime at this level. As a result of exciton annihilation, new pairs of photons appear. One of the pair of photons leaves the medium—a contribution to PL. The second throws an electron to one of the higher levels of the alloying atom. The non-radiative relaxation of such an electron to the lower levels is the reason for the heating of the medium.

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