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The role of Bohr frequencies in the scattering, luminescence, and generation of radiation in different media

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Contents

Abstract. The Bohr correspondence and complementarity principles, which express the endeavor to utilize the concepts of classical theories (mechanics, electrodynamics) to the highest degree, are invoked in this paper to describe the interaction of light photon beams with a medium whose properties can be represented using the Lorentz model of the classical harmonic oscillator and the dispersion theory.

1. Introduction

In the consideration of the atomic model, Niels Bohr came up with three quantization postulates. I mention, following Tolanskii [1], the contents of these postulates. The first consists in the fact that there exists a certain number of electron orbits in an atom, which Bohr termed stationary states. Moreover, an electron may reside in each of these states for an arbitrarily long time without radiating at all. This proposition was arbitrarily postulated, and it was certainly at variance with the classical electromagnetic theory. Second, Bohr postulated that an electron can be knocked out of the lower stationary, normal state (E_1) to a higher-energy state (E_2), which he termed an excited state. The transition between these states is characterized by the Bohr frequency [2], whose value is defined by the relation

$$\nu_{21} = \frac{E_2 - E_1}{h}\,, \tag{1}$$

where h is the Planck constant and $E_2 - E_1 = \Delta E$. Third, he assumed that in the stationary orbit, the quantity termed the electron angular momentum should be proportional to the Planck constant. Using the above assumptions, Bohr arrived at the theoretical Balmer formula, derived the exact values of the Balmer line wavelengths, and predicted the existence of other series. Bohr's ideas proved to be beneficial for the explanation of spectra and found use in practical spectroscopy.

The present paper considers the scheme of the photon beam interaction with resonance and near-resonance media, whereby the exciting radiation frequency ν at the input to the medium and the frequencies of the outgoing radiation at the output are unambiguously related to the Bohr frequencies [see relation (1)] both in the case of exact resonance $\nu = \nu_{21}$ and in nonresonance conditions $\nu < \nu_{21}$, $\nu > \nu_{21}$. The practical application of the proposed approach turns out to be helpful in the consideration of amplification, luminescence, and scattering of radiation in different (atomic, molecular, nanodimensional) media with a three-dimensional distribution of oscillators.

In describing the elementary processes of the radiation – substance interaction (absorption, spontaneous and stimulated emission) in this paper, we more frequently resort to the term photons, not forgetting about the distinction between the notions of light quanta and full-value photons drawn by Ginzburg [3].

2. The Bohr frequency and its relation to the Lorentz model of a classical harmonic oscillator

The concept of nonradiative relaxation transitions, also related to two levels of a medium by formula (1) for the Bohr frequency, emerged in atomic physics even prior to the advent of lasers [4]. As a rule, these transitions were due to the interactions between the excited particles (atoms, molecules, mesoatoms, and nanoparticles) of the medium and their environment. These interactions may be regarded as collisions between the particles and their environment, depending on the type of specific system. In the long run, the result of the nonradiative relaxation transitions is the energy exchange between the subsystem of the particles involved and the thermal motion in the entire system, which leads to equilibrium in the system as a whole.

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The equilibration time, or the particle lifetime in a level, is usually denoted by T_1 and is called the longitudinal relaxation time [4]. The quantity $1/T_1$ is the rate with which the population difference between two states related by condition (1) returns to its equilibrium value as a result of energy transfer from the particle ensemble to the thermostat [4, 5]. According to Ref. [5], both spontaneous emission [3] and nonradiative relaxation make a contribution to T_1. The total time an atomic ensemble takes to completely lose the excitation energy is, as a rule, close to T_1. This proposition is partly at variance with the first Bohr postulate, which states that a closed electron orbit must be stable and that the electron residing in this orbit does not radiate energy and would stay in the orbit (level) for an indefinitely long time.

It is noteworthy that spontaneous emission as well as nonradiative relaxation can occur when the light field acting on the particles of the medium has already been turned off. In the presence of a resonance light field, of significance are the processes of stimulated emission and (stimulated) absorption of the light field quanta.

When the density of a medium is high and the field is absent, the role played by radiative transitions is negligible in comparison with the nonradiative ones and the medium can stay in the thermodynamic equilibrium. This implies that the atoms obey the Boltzmann distribution [6] over the levels:

$$\frac{n_2}{n_1} = \exp\left(-\frac{\Delta E}{kT}\right), \tag{2}$$

where k is the Boltzmann constant, T is the temperature, and n_1 and n_2 are the respective numbers of electrons in the lower and upper transition levels.

Exposure of a medium to the light of an external source can disturb the thermodynamic equilibrium of the medium. In this case, the relation between the spontaneous emission probability and the probabilities of absorption W_{12} and stimulated emission W_{21} is characterized by a factor that involves the spectral intensity of the incident radiation or its spectral density with the constraint that the probabilities of stimulated emission and absorption are equal [3, 6].

The problem of the role of nonradiative relaxation in the processes under discussion was investigated by Karlov [4] on the basis of the analysis of population variation in a two-level system exposed to a resonance electromagnetic field. According to Ref. [4], the variation in the particle number density n_2 in the upper level is given by

$$\frac{\mathrm{d}n_2}{\mathrm{d}t} = -\left(w_{21} + \frac{1}{t_1}\right) n_2 + w_{12} n_1 - W_{21} n_2 + W_{12} n_1\,, \tag{3}$$

where w_{12} is the probability of a spontaneous electron transition from the ground level to the excited one, w_{21} is the nonradiative relaxation probability, and $1/t_1$ is the rate of particle escape from the second level due to spontaneous emission.

It is significant, and this is the central point of the present paper, that irrespective of whether the ensemble has completely lost the excitation energy, the medium permanently retains the memory of the existence of a transition between the levels E_2 and E_1 and of the value of the Bohr frequency ν_{21} [see relation (1)]. In other words, the Bohr frequency is an invariable characteristic of the medium under investigation, irrespective of whether the electron occupation of the transition levels is equilibrium or nonequilibrium. Unambiguously related to the Bohr frequencies are the oscillators (as a rule, these are the optical electrons of atoms, molecules, mesoatoms, and nanoparticles) distributed over the volume of the medium. The eigenfrequencies of these oscillators are determined by elastic forces and are independent of the external field frequency [7].

It is pertinent to note that in the absence of exact resonance between the excitation field frequency ν and the Bohr frequency ν_{21}, which characterizes the two-level system under study, stimulated electron transitions are effected involving multiphoton processes, specifically, three-photon electron Raman scattering (RS) [8 – 10] and six-photon parametric scattering (SPS) [10, 11]. We recall that an elementary event of the three-photon process involves the absorption of two photons of the exciting radiation at the frequency ν and the emission of a photon at the combination frequency ν_3 (in the literature, this process is sometimes called hypercombination or Raman [12] scattering). In the case of a two-level medium characterized by the Bohr frequency ν_{21}, the frequency ν_3 of the three-photon electron RS is, in accordance with the energy conservation law, expressed as

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

(the factor h is omitted).

Several research groups have reported the experimental observation of this process in Refs [8 – 10], which give bibliographic references to three-photon scattering. For information about its theoretical treatment, the reader is referred to Ref. [13].

In the case of SPS in a two-level medium, according to the energy conservation law, the three absorbed photons of exciting radiation correspond to the production of three photons at new frequencies. The process should not violate the equilibrium conditions in the medium. In this case, of significance is the fulfillment of the momentum conservation law for the six photons participating in the elementary interaction event. This process was considered in Refs [10, 11].

3. The correspondence principle as an endeavor to make full use of the notions of classical theories

We now turn our attention to another point made by Niels Bohr [14]. The point at issue is the complementarity and correspondence principles. According to Bohr, there exists the same relation of complementarity between the unequivocal application of stationary states and the mechanical analysis of intraatomic motion as between a light quantum and the electromagnetic radiation theory. The correspondence principle expresses the endeavor to harness the concepts of classical theories (mechanics, electrodynamics) to the highest degree, despite the contradiction between a light quantum and the electromagnetic theory of radiation [14].

The aim of this paper is to interpret, in accordance with the Bohr correspondence principle, the features of the spectral structure of the medium-scattered (secondary) radiation excited by the beams of resonance or near-resonance light quanta (photons) and to search for the universal rules for processes of this kind with the use of the notions of the classical theory that pertain to the resonance medium. As shown below, using the processes that correspond to relation (4) or the like (see Section 4) promotes a clear classification and interpretation of the spectra of in-medium-scattered radiation and luminescence

spectra, as well as light amplification. The proposed model enables a valid explanation of several features of the resonance and near-resonance interaction of monochromatic radiation with different (atomic, molecular, nanodimensional) media.

In an endeavor to make the fullest use of the notions of classical theories, we mention that the mechanical Lorentz model of a classical harmonic oscillator exists precisely for the description of the properties of an atomic (molecular) medium [15].

Having discussed the Bohr frequency, the relation of complementarity, and the Bohr correspondence principle, we now recall that the electrons responsible for the optical properties of a medium may be represented as an ensemble of similar or variously sized classical harmonic Lorentz oscillators, which in turn requires invoking the results of the dispersion theory [15, 16] related to these oscillators. Our prime concern is with how a photon beam of monochromatic radiation of frequency ν interacting with a nearly resonance medium can propagate through a medium that obeys the dispersion laws. In the material media under investigation, an ensemble of classical harmonic oscillators corresponds to every Bohr frequency ν_{21}.

Our interest is with three different situations:

A) $\nu = \nu_{21}, n(\nu) \simeq 1$,

B) $\nu > \nu_{21}, n(\nu) < 1$,

C) $\nu < \nu_{21}, n(\nu) > 1$,

where ν_{21} is the resonance transition frequency, which naturally corresponds to the Bohr frequency [see relation (1)], and $n(\nu)$ is the refractive index of the medium.

In case A, according to the dispersion theory, the refraction index $n(\nu)$ of the medium with a volume oscillator distribution is close to unity [15, 16]. According to Ref. [15], the reflection coefficient of this medium peaks at the resonance frequency. When a population inversion is produced in the medium at the transition with the frequency ν_{21}, radiation amplification is bound to occur.

In case B, a special feature limiting the photon propagation is related to the fact that the refractive index $n(\nu) < 1$, according to the dispersion theory. The propagation of photons of monochromatic radiation turns out to be impossible in this spectral region, because otherwise they would propagate with a supraluminal speed $v = c/n(\nu)$ (c is the speed of light in empty space), which is at variance with the existing notions. Only by effecting the saturation of the medium, i.e., the level population equalization of the transition under investigation, is it possible to transmit the radiation through this medium. Otherwise, according to Ref. [15], efficient reflection of the incident radiation occurs at the boundary of a medium with a volume distribution of classical Lorentz oscillators.

Finally, in case C, according to the Sellmeier relation [16], an unbounded growth of the refractive index $n(\nu)$ occurs in the low-frequency spectral region relative to the resonance frequency. This circumstance may be responsible for a very strong moderation of the photons that penetrate the medium and may therefore impede their propagation.

4. Several experimental facts and their discussion on the basis of the proposed model

The problem of the resonance and near-resonance interaction of radiation with different media has attracted the attention of researchers for a long time (Wood, Rozhdenstvenskii).

The systematic study of resonance interaction became possible with the advent of frequency-tunable cw or pulsed lasers. Analyzing the experimental results in this area of research might seem impossible due to the extensive body of publications. My task is to extract those results from the wealth of experimental data which correspond to the multiphoton model considered in the present paper. For this, I use the published materials obtained with my participation, as well as the findings of other researchers available in the literature.

We enlarge on the experimental data that serve to illustrate case A. Here, it is pertinent to recall Wood's classical experiments (see Ref. [17]) in which he observed the opacity of a medium (sodium vapor) to the incident resonance radiation. With an increase in the density of the atomic sodium vapor, the region of vapor glow shrinks to the point of beam entry into a vapor-containing cell and, in the view of the observer, turns into the glow of a thin atomic surface layer (or into the reflection of light by an atomic surface layer). That is, the forward propagation of monochromatic radiation at the resonance frequency ($\nu = \nu_{21}$) is limited. According to Ref. [18], the opacity (strong attenuation) of the medium to resonance radiation can be attributed, from the standpoint of classical electrodynamics, to the high magnitude of the scattering cross section σ. In the case where the frequency of exciting radiation coincides with the Bohr frequency of the medium ($\nu = \nu_{21}$), the scattering by bound atomic electrons limits the directional radiation propagation through the cell with an increase in density of the alkali metal.

When a two-level inverted system is irradiated by photons whose frequency ν is equal to the Bohr frequency of the medium ($\nu = \nu_{21}$), the scheme of stimulated (induced) emission of photons with the same frequency ν is realized, which was proposed by Einstein in 1916 [19]. The production of two photons in an elementary event in an inversely populated resonance medium with the simultaneous absorption of one photon corresponds to the expression

$$\nu + \nu'_{21} = 2\nu\,, \tag{5}$$

identical to equality (4), where $\nu = \nu'_{21}$ and $\nu'_{21} = (E_2 - E_1)/h$ is the Bohr frequency of the inverted transition of the two-level medium under investigation. Relation (5) uncovers the mechanism of light radiation amplification involving the doubling of the number of photons in every elementary event. This process has found wide application in many problems of quantum radiophysics and electronics related to the amplification and production of coherent monochromatic radiation [4, 12]. Interestingly, even at the dawn of the age of laser physics, the model of the classical harmonic Lorentz oscillator that we invoke was employed in the discussion of lasing [20].

We now turn to the analysis of experimental data that serve to illustrate case B: $\nu > \nu_{21}, n(\nu) < 1$.

From the situation whereby the propagation of photons at the frequency ν through the medium is limited in the specified spectral range, it is possible to find a way out by assuming the possibility of the production of a precursor [15], which testifies to a change in the properties of the medium for the photons that follow behind the leading edge of the beam. The experimental data on the interaction of nearly resonance, monochromatic radiation with atomic potassium vapor carried out with the participation of the author of the present paper [8, 10, 11, 21, 22] allow concluding that in the case of a resonance (two-level) medium, the role of the precursor

should be assigned to the photons whose frequency ν_3 corresponds to the above relation (4): $\nu_3 = 2\nu - \nu_{21}$, where ν_{21} is the Bohr frequency of the medium. Because $\nu_3 > \nu$ and $\nu > \nu_{21}$ in this case, we have $n(\nu_3) > n(\nu)$. Furthermore, according to the dispersion theory [15, 16], we expect that $n(\nu_3) \to 1$ as the frequency increases, which removes the limitation on the radiation propagation.

The observation of such processes in nearly resonance conditions in atomic alkali-metal vapors was reported by other researchers in Ref. [9]. Reference [9] contains a bibliographic list of mainly experimental works dedicated to the observation of three-photon process (4) in atomic media.

Indeed, as a result of process (4), equilibration of the level populations of the medium investigated occurs (electrons are transferred from the ground level 1 to the excited level 2), which testifies to the formation of conditions in the medium for the unimpeded transmission of the radiation at the frequency ν_3 and immediately after at the frequency ν. The radiation with the frequency ν_3, which is successfully recorded on spectrograms, is indication that the process is appreciably intense [8, 9].

The illusory contradiction that follows from the smallness of the estimated cross section σ_3 of the three-photon process given in Ref. [12, p. 173] and its accessibility for observations in atomic alkali-metal vapors [8, 9], is attributable to the fact that the lower-order processes in a medium with a refraction index $n(\nu) < 1$ are insignificant: they do not mask the radiation at the frequency ν_3.

The self-focusing of laser beams with a Gaussian intensity distribution over the beam section, which is observed in alkali-metal vapors [21–23] in this spectral region, according to the model under discussion, may be represented as the tunneling regime [22] for the photons with the frequency ν through the channel prepared by process (4). In conformity with this process, the first to appear on the beam axis as the precursors are photons with the frequency ν_3.

We next analyze case C ($\nu < \nu_{21}$, $n(\nu) > 1$) with recourse to available experimental data. First, we consider a medium consisting of two-level atoms (by the example of atomic potassium vapor [10, 11, 22]) and, second, the luminescence observed in nanodimensional media [with the proviso that $\nu < \nu_{m1}$ and $n(\nu) > 1$, where ν_{m1} are the eigen (Bohr) frequencies of the nanodimensional oscillators of the medium; $m = 2, \ldots$] [24, 25]. According to Refs [24, 25], the conditions required in case C [$\nu < \nu_{m1}$, $n(\nu) > 1$] in the presence of luminescence are automatically satisfied for the set of Bohr frequencies ν_{m1}, which are located, as a rule, in the blue (anti-Stokes) spectral region relative to the frequency ν of the exciting radiation. The luminescence spectrum itself is shifted to the Stokes spectral region relative to ν [4, 26].

It is pertinent to note that in the case of potassium atoms, we are dealing with calibrated oscillators and an invariable Bohr frequency for a given transition. In the case of the luminescence of a nanodimensional (or molecular) medium, due to the different dimensions of the nanoparticles we obtain a set of Bohr frequencies ν_{m1} ($m = 2, \ldots$).

1. Therefore, for a medium of two-level atoms, expression (4) for the three-photon electron RS is of the form $\nu_3 = 2\nu - \nu_{21}$, where the quantity ν_{21} corresponds to the Bohr frequency of the transition under investigation, ν is the exciting radiation frequency, and $\nu < \nu_{12}$. In this case, according to Refs [15, 16], $n(\nu) \gg 1$, which should lower the radiation propagation velocity in this spectral region. For laser radiation beams with a Gaussian intensity distribution over the beam section, it has been possible to observe the Vavilov–Cherenkov effect in alkali-metal vapors in this spectral region. This effect is caused either by the supraluminal propagation of the nonlinear polarization induced in the medium [10] or by the propagation of supraluminal photons through the resonance medium [11], which actually travel with a speed that does not exceed the speed of light c. Owing to process (4), which takes the Bohr frequency into account, a channel in which $n(\nu) \to 1$ forms on the axis of the light beam due to population equilibration, which furnishes the conditions for Cherenkov radiation [27] with the use of this unconventional supraluminal source.

In considering cases A, B, and C, it has been possible to show that it is precisely the Bohr frequencies of a medium with a volume distribution of classical harmonic oscillators that define the properties of interaction of monochromatic radiation beams with such media. For similar calibrated oscillators of the medium (atomic alkali-metal vapor), the experimental results are evident and are easily amenable to interpretation. The process of nonradiative relaxation ensures dissipation of the energy stored by the oscillators of the medium due to process (4). We note that the Bohr frequencies themselves are, as a rule, not recorded on spectrograms except in the case of exact resonance (5), whereby $\nu = \nu'_{21}$.

Therefore, experimental investigations into the propagation of laser radiation beams in a two-level atomic medium confirm that process (4) plays the decisive role in the interaction of laser radiation with the nearly resonance two-level medium.

2. The situation is more complicated for a medium with a volume distribution of oscillators with different dimensions. In this case, the spectrum of Bohr frequencies is broader, entailing a complication of the radiation spectra scattered by the medium.

We address ourselves to one more phenomenon, specifically to the effect of fluorescence (fast luminescence) [26], and consider the consequences of applying the proposed model to this process. The reason for this approach is as follows: until recently [24, 25], the relation between the observed spectra and the specific oscillators of the medium was not taken into account in the interpretation of the above effect. We try to establish this relation. Relation (4) is rewritten as

$$\nu_m = 2\nu - \nu_{m1}\,, \tag{6}$$

where $m = 2, \ldots$ denotes the number of one of the numerous frequency spectral components of the luminescent emission, which consists of the frequencies ν_m and is scattered into an angle of 4π steradians in a medium with a volume oscillator distribution, and $\nu_{m1} = (E_m - E_1)/h$ are the characteristic Bohr optical frequencies for electrons (of a complex organic molecule or a nanodimensional particle) embedded in one or other environment. This environment is, as a rule, size-noncalibrated aggregates of different dimensions consisting of some combination of atoms, molecules, or fragments of a crystalline structure with numerous defects. In accordance with the problem formulated, we should show how the Bohr frequencies ν_{m1} of suchlike aggregates are related to the luminescence spectrum. As in the case of a two-level atom, we reason about the necessity of borrowing a part of the medium-exciting radiation to effect the dynamic compensation of the dispersion of refractive indices of individual oscillators in the medium, which turn out to have different dimensions in this case [$n(\nu_{m1}) \to 1$]. In our view, relation (6) describes this process, as evidenced by the luminescent

emission at the frequencies ν_m. In the recently published papers [24, 25] of the present author, the luminescence of silicon nanoparticles suspended in ethanol is described in detail employing the present approach.

Upon discussing the problems related to resonance and near-resonance media and the Bohr frequencies, we have to revert to relation (3) considered in Section 2. The relevant case pertains to the absorption W_{12} and stimulated emission W_{21} probabilities. Clearly, these quantities should take the three-photon processes (4) – (6) considered above into account [11].

5. Conclusion

We have considered several practical problems of scattering, light amplification, and luminescence in resonance or nearly resonance media with a volume distribution of classical oscillators. The eigenfrequencies of these oscillators correspond to the complete set of Bohr frequencies of the media under investigation. The properties inherent in the interaction of light quanta beams with such media are described. Irrespective of whether the conditions for exact resonance between the exciting radiation frequency and the Bohr frequency of the medium are satisfied, the participation of three photons in an elementary radiation – medium interaction event ensures the ground-to-excited (or vice versa) oscillator state transition. The correspondence principle and the Bohr principle of complementarity, which express the endeavor to utilize the concepts of classical theories (mechanics, electrodynamics) to the highest degree, in the above-considered example are directly related, on the one hand, to the quantum properties of light radiation and on the other hand to the mechanical properties of the classical harmonic Lorentz oscillator and the dispersion theory.

Using relations (4) – (6) facilitates the identification of the spectral structure of the radiation scattered by a resonance or nearly resonance medium in different directions. On the one hand, the question arises of how closely the exciting radiation frequency ν can be brought into coincidence with the frequency ν_{21} or ν_{m1} of the transition under investigation. On the other hand, in the case of a so-called nearly resonance interaction (both for two-level atoms and for luminescence), it is not necessary to be concerned about the exact coincidence of the frequency ν with the Bohr frequency ν_{21}. Recording the output radiation at the frequency ν_3 (ν_m) in an experiment with a resonance or nearly resonance medium supports the proposed scheme. It is noteworthy, however, that the experimental detection of radiation with the Bohr frequency ν_{21} or ν_{m1} in the absence of exact resonance, as a rule, turns out to be difficult or impossible, for instance, due to nonradiative relaxation. Experimental data nevertheless allow calculating this frequency.

Relations (4) – (6), which are borne out in numerous experiments on the amplification and scattering of laser radiation, the interaction of laser radiation with nearly resonance two-level media, and the fluorescence of nanodimensional particles, have a universal nature in our view. There is good reason to apply this law for the interpretation of experiments in atomic, molecular, and nanodimensional media. Special emphasis should be placed on the role played by the Bohr frequencies ν_{21} (ν_{m1}), which are integral characteristics of the medium under investigation. In particular, using this rule in fluorimetric analysis provides an independent expert evaluation of the characteristic frequencies of the intrinsic oscillators of the medium investigated. Reverting to the notion of elementary classical oscillators of the medium, which are responsible for the Bohr frequencies, seems to be expedient in the study of the above-listed media and processes, because it enables gaining new information about the characteristics of the medium.

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