Physical origin of nonlinear phenomena in optics.

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Abstract

The physical nature of numerous of the nonlinear phenomena in optics is explained by inequality of forward and reversed optical transitions, that corresponds to a principle of time invariance violation in electromagnetic interactions. The direct and indirect experimental proofs of such inequality and direction of the further researches are discussed.

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1 Introduction

The research of the physical phenomenon usually pursues two purposes. The first purpose is to explain qualitatively a nature of the phenomenon and the second one is to describe it quantitatively with the help of suitable mathematical model. These tasks frequently have a different degree of complexity and its not always can be executed simultaneously. It happens, that the explanation of physical effect is rather simple, and its quantitative description is very difficult. It is a case with a prediction of weather.

There are cases when the phenomenon appears easier to describe quantitatively, than to understand its physical nature. The periodic law is such vivid example. It is the main law of chemistry. The periodic table was created by Mendeleyev on the base of atomic weights. It gives excellent description of elements and their compounds properties. But this law for very long period had not a physical explanation. Only there are a lot of years later, after discovery of electrons, protons, neutrons and creation of the quantum mechanics, the physical essence of the law has become clear.

The situation, similar to this case, probably, has a place now in optics. Many years the mathematical description of dynamics of optical transitions exists. It is based on the Bloch equations, which was offered in 1946 for the description of a nuclear magnetic resonance [1]. Now Maxwell-Bloch equations are used for the description practically all nonlinear effects in optics. Such model gives really good description of the optical phenomena [2, 3]. However the Bloch equations have not clear physical interpretation. Therefore there are large difficulties at attempts to understand physical sense of such descriptions. For this purpose such concepts, as coherence, interference and a superposition of states are usually used. But its nothing explain. Its look like as attempt of physical interpretation of the mathematical description, when to operations
of addition and subtraction of the members in the equations there correspond principles of interference and states mixing.

In present article the physical explanation of set of the phenomena in optics is discussed. It is based on a principle of inequality of forward and reversed optical transitions. However, for a long time and strongly settled opinion exists that such optical transitions are equivalent, and electromagnetic interactions as a whole in nature are time invariant [4]. It is difficult to understand what is the base of this opinion. Usually it is referred to Einstein’s opinion ”that physics could be restricted to the time-symmetric case for which retarded and advanced fields are equivalent” [5].

The basis of this opinion can not be the equality of Einstein coefficients for absorption and stimulated emission of photons. Einstein coefficients characterize the integrated cross-section of optical transition. The preservation of time invariance demands not only equality of integrated cross-sections, but also equality of spectral width of direct and reversed optical transitions. Thus, the Einstein coefficients have not the direct relation to invariance of process of photon absorption.

The experiments on study of differential cross-section of reaction $\text{Al}^{27} + p = \alpha + \text{Mg}^{24}$ also can not be considered as the proof of time invariance preservation [6]. These experiments show equality of differential cross-sections of forward and backward processes. But we are interested more in comparison of cross-sections forward and reversed processes. It is a case, when the backward process follows directly after the forward one and the system can have memory about the forward process. So, the experimental proofs of time invariance preservation in electromagnetic interactions are absent.

From other side the opinion exists, that (except for the case of $K^0$-meson decay) there are absent also the experimental proofs of time invariance violation [7]. This is error. In optics there is a set of the experimental proofs of time invariance violation in electromagnetic interactions. But usually there are the indirect experimental proofs, which essence is not realized to the present time.

The problem is that we are not able to measure separately the parameters of forward and reversed processes. Under action of radiation both forward and reversed processes proceed simultaneously. A width of optical transition is usually connected with lifetime of the exited state. Therefore, when wide optical transitions are used, some temporary difficulties appear. If the lifetime of exited state is big, the line of optical transition is very narrow and difficulties with inhomogeneity of a spectrum exist. As a result the information on parameters of the reversed optical process has mainly indirect character. For example, the Autler-Townes effect [8] and the numerous examples of amplification without inversion [9] are the indirect proofs of inequality of forward and reversed processes.

However the brightest indirect proof of this inequality in our opinion is the effect of adiabatic population transfer in two-level system due to sweeping of resonant conditions [10]. When the resonance radiation interacts with the two-level system, the so-called periodical Rabi oscillations of the level population takes place. But if the sweeping of resonance conditions appears (for example,
the frequency of radiation is changed), the full population transfer from the
initial level to the opposite one takes place. And this result does not depend
on intensity of radiation (if it is rather strong). This surprising result is well
described in Bloch model. It is said, that the physical nature of this effect can
hardly be explained verbally in simple terms, but one should carefully follow the
behavior of the vectors in the model of the rotating wave [11]. This situation
resembles very much an appeal of the prestidigitator asking the spectators to
watch his hands carefully while he is making the manipulations. And the result
seems to be the same. An explanation is given, but its physical essence is
absolutely unclear.

Here it is important that the physical explanation of this effect is impossible,
if we assume equality forward and reversed processes. Its should be something
different, that the atom could know, what level is initial, and what level is final.
It is natural to expect, that the difference can consist in different width and
cross-section of forward and reversed transitions. The rapid adiabatic passage
effect is the most convincing indirect proof of inequality forward and reversed
processes.

However for the decision of a problem only indirect proofs are insufficient.
The direct proofs are necessary. And such direct proof exists. It is connected
to physical object, which has an unusual combination of properties: extremely
large homogeneous width of optical transition is combined with the big life-
time of the exited state toward to spontaneous emission. In this case it appears
very easily experimentally to find out the large difference between parameters of
forward and reversed processes. This unusual object is the so-called wide com-
ponent of line in absorption spectrum of polyatomic molecules. The following
section is devoted to discussion of experiments with this object.

2 Infrared laser multiple photon excitation of
polyatomic molecules.

The phenomenon of the infrared multiple–photon excitation (IRMPE) and
collisionless dissociation of polyatomic molecules was discovered in works [12,
13]. It was founded, that polyatomic molecules can absorb tens photons of
laser radiation and dissociate without collisions. Numerous works were carried
out later aimed to clarify the mechanism of this process. This interest was
stimulated by the fact, that the widths both of laser radiation and molecule
absorption lines are substantially lower, than the anharmonicity of molecular
vibrations. It means, that the absorption of second quantum of laser radiation
should not occur.

For an explanation of the mechanism of process in the former work the
hypothesis about existence of so-called ”quasicontinuum” of vibrational states
was proposed. Despite of the argued criticism [14] such idea has received the
broader distribution. In the recent years, however, the views on the nature of ”quasicontinuum” have changed dramatically. Earlier, it was accepted, that
"quasicontinuum" consists of a huge number of narrow lines arising as a result of coupling different vibrational states.

Now it is widely believed [15], that the absorption line is unique, but it becomes very wide. The origin of the "quasicontinuum" now is bound up with intramolecular vibrational relaxation (IVR) process. This is a reasonable idea. The IVR process can be very fast (picoseconds timescale). The corresponding Lorentzian width of the absorption line can be in this case comparable with anharmonicity of the molecular vibrations. The main disadvantage of this model is that it does not explain how the molecules can be excited in the region of low vibrational levels, where the IVR is absent and the absorption lines remain narrow. Experiments show, that excitation of molecules in this area occurs without essential difficulties, but the theory gives no satisfactory explanation of this fact.

In works [16, 17] an idea was proposed, that the IR MPE process is a trivial result of absorption in the area of line wings, but antiviral is the nature of these wings. Practically, the possible role of line wings was not discussed in the literature earlier. It is, apparently, due to the fact, that appropriate estimations can easily be made. The lifetime of excited states of molecules due to spontaneous emission in the infrared region lays in the millisecond timescale. The natural width of line must to be smaller than 100Hz. Even for the strongest molecular transitions, at the distance from the line center equal to the value of molecular anharmonicity, the Lorentzian contour of the natural width would have an absorption cross-section smaller then $10^{-25}$ cm$^2$. This cross-section cannot play any appreciable role in overcoming the anharmonicity of molecular vibrations.

However, such estimation has not been tested in experiment earlier. It is possible to assume, that for some unknown reasons, intensity of real line wings is much higher, than the theory predicts. How high the intensity of line wings should be to explain the observable effect of laser excitation of molecules? Rather correctly such information can be derived from the experimental results of works [18-20], where the depletion of rotational states of SF$_6$ molecules by TEA CO$_2$–laser radiation was studied in the conditions of molecular jet. The results of such processing are presented in Fig.1. Except for the usual narrow component of the line with a Doppler width $\sim 30MHz$, the wings, or more precisely speaking, a wide component of the line should exist with a cross-section $\sigma \approx 6 \cdot 10^{-20}$ cm$^2$ and with a Lorentzian full width at half medium $\sim 4.5$ cm$^{-1}$. The relative integral intensity of this component is rather small, $\sim 0.2\%$, but it is high enough for efficient excitation of molecules from all rotational states.

For experimental test of existence the wide components of a line the form of line should be studied on the large depth. This strongly prevents by inhomogeneous broadening, which connected with distribution of molecules on different rotational states. At room temperature the dense spectrum of transitions from different rotational states is observed. But here it is important to pay attention that Lorentzian contour is rather flat and wide component of lines can manifest itself as far natural wings of absorption bands. There are many publications about study of far wings of absorption bands of small and light molecules [21,22]. These wings are the result of collisional broadening of
absorption lines. For heavy polyatomic molecules in a gas phase the far wings of absorption bands of other nature were discovered [23]. The experiments have shown, that the cross-section of absorption in the region of these wings does not depend on pressure of gas. So, its have a natural nature.

In Fig.2, the spectral dependence of the absorption cross-section of $\text{SiF}_4$ molecules around the $\nu_3$ absorption bands is presented. The edges of the absorption band have approximately an exponential form, the slope being greater for the blue side, than for the red one. At the distance more, than $25—40 \text{cm}^{-1}$ from the band center, much more flat wings are observed. The curve (2) is a Lorentzian profile with FWHM $= 4.5\text{cm}^{-1}$, which passes through the point with minimal absorption cross-section in the given spectral range. So, we can see, that the far band wings have a Lorentzian behavior.

In Fig.3 a spectral dependence of the absorption cross-section of $\text{SF}_6$ molecules is shown. In this rather typical case the far band wings are masked by intense combination bands.

The measurement of intensity of far absorption band wings allows to estimate integrated intensity of wide components of lines. The same value can also be experimentally estimated by other method: on the data about saturation of absorption spectrum of polyatomic molecules by radiation of pulse $\text{CO}_2$-laser at low gas pressure [23]. The experiments show, that the relative intensity of wide component quickly grows with increasing of number of atoms in a molecule and branching degree of the molecules. Thus the estimated average relative integral intensity of the line wings at room temperature varied from $\sim 0.6\%$ for $\text{SF}_6$ and $\text{SiF}_4$ to $\sim 90\%$ for $(\text{CF}_3)_2\text{O}$ and $(\text{CF}_3)_2\text{CO}$.

The nature of wide components of lines is unknown. As a hypothesis the following explanation is offered. A certain mechanism of averaging of the rotational moment of molecule inertia works during the vibrational motion of atoms. In the large molecules this mechanism undergoes periodic and convertible breaking. As a result the absorption line splits on a clump of narrow lines, and the short-lived moments of breaking correspond to a wide component of a line [24].

The wide component of line is unique physical object, which has the long lifetime of the excited states toward to spontaneous emission and large homogeneous spectral width of optical transition. This combination of properties is very convenient for study the reversed optical transition in conditions of a molecular beam.

A wide component of lines was easily observed in work [25] at study the absorption of radiation of continuous $\text{CO}_2$- laser in a molecular beam with cryogenic bolometer. Rotational temperature in a molecular beam is very low ($\sim 5^0\text{K}$). It radically changes character of a molecule absorption spectrum. The absorption lines become very rare and the $\text{CO}_2$- laser radiation interacts practically only with the wide component of lines. Unfortunately, the authors had not understood with what thing they deal with and later the work with this object was closed.

In present case we are not interested in a line wings itself, but in the results of double optical resonance experiments in a molecular beam [25]. For the first laser beam the absorption spectrum represents wide continuum. For the second
laser beam, which cross the molecular beam later, besides this wide continuum a sharp dip with a width $\sim 450\text{kHz}$ is observed. It characterizes a spectrum of the reversed optical transition. The ratio of forward and reversed optical transitions widths exceeds $10^5$ times for the given case.

Besides this, the amplification of probe laser radiation was observed. Taking into account, that in some of these experiments the number of the molecules, exited by the first laser, did not exceed $\sim 0.1\%$, it is a typical case of amplification without inversion. Thus the cross-section of the reversed optical transition should be at least on three orders of magnitude more, than of forward one. Because of the Einstein coefficients for forward and reversed transitions should be equal, the present estimation for cross-section of the reversed transition is, obviously, underestimated.

Now these experiments are the unique direct and complete experimental proof of inequality of forward and reversed optical transitions. However, the given physical object, probably, is not so unique. The rather similar experimental results were received earlier in a solid state. In homogeneously broadened absorption line of a ruby the dip with width only $37\text{Hz}$ was observed [26,27]. And in this case the huge homogeneous width of optical transition is combined with the big lifetime of the exited state toward to spontaneous emission ($\sim 3,4\text{ms}$). The measurement of width of the dip was carried out by amplitude modulation of laser radiation. Unfortunately usual pump-probe experiments with using two pulse lasers were not carried out. Such experiments have not so high spectral resolution. But such results can have more evident, convincing and direct physical interpretation.

3 Nonlinear optical effects.

The traditional explanation of a nature of nonlinear optical processes has descriptive character. Their existence are connected with so-called nonlinear susceptibility of atoms and molecules [28]. If appropriate factor of a nonlinear susceptibility is great enough, the process, for example, of a wave mixing can occur. If this factor is equal to zero, the wave mixing is absent. Also, a restriction on the lowest order wave mixing process exists, that is connected with the symmetry of the process. Namely, the factor of nonlinear susceptibility $\chi^2$ is nonzero only in a medium without center of inversion.

Within the framework of discussed representations the main reason of nonlinear optical processes is the inequality of forward and reversed optical transitions. As the result the atoms and molecules have some "memory" about their initial state and aspiration to return back. A basis of this aspiration is the high cross-section of optical transition into the initial state. And it is necessary to understand that the concept of initial state should include not only concrete quantum energy level, but also the orientation of atom in space toward the laser beam and even a phase of vibrational motion of atoms in a molecule.

In this case it becomes clear why in gas and liquid phases there are no effective processes of three and five photon mixing. Every photon has spin.
Therefore, using odd number of photons it is impossible to return to an initial state. At the same time in crystal lattice, where the rotation can be suppressed, such processes are possible.

But in gas and liquid phases there are a huge amount of four photon mixing processes. The set of nonlinear optical effects are possible and necessary to consider as result of four photon mixing process [29].

Here we shall discuss in detail only one such effect. It is one of the most complex effect. Despite of old history and constant interest to it, it is insufficiently well experimentally investigated till now. The main problem here is that the existing theoretical representations about its nature strongly prevents experimenters. There is a photon echo effect.

The atoms and molecules, exited by laser radiation, are capable to radiate coherently this energy. When it occurs at once after laser pulse, such radiation calls as a nutation effect. The complete radiation of the reserved energy does not occur. The dephasing processes are one of the reasons. Some of the dephasing processes can be reversed in time, then a pulse of a photon echo superradiation can appear [2].

Usual two-pulse photon echo arises when environment is influenced by two pulses of laser radiation (P1 and P2) with a delay $t_{21}$ between them. Then at time $t_{e2} = t_{21}$ after the second laser pulse there is a pulse of photon echo. There is also variant of stimulated photon echo, when on environment work with three laser pulses (P1, P2, P3). Then the pulse of photon echo occurs after a pulse P3 with a delay equal delay between pulses P1 and P2 ($t_{e3} = t_{21}$). The time of a delay $t_{32}$ in this case can be very large.

The Bloch equations well describe dynamics of photon echo, but nothing says about a physical nature of this process. The concepts of optical, Zeeman coherences and superposition of states are used for explanation a photon echo effect [30]. However, such approach contradicts some experimental facts. So the study of stimulated photon echo in a case, when the delay time $t_{32}$ between the second and third pulses considerably exceeds the lifetime of the excited state of atoms, clearly specifies that the information about the photon echo in this period of time contains only in a unique ground state [31]. In this case such result is a natural consequence of small lifetime of the excited state. But such data can be received also in other way. Using an additional laser pulse of other frequency [32], it is possible to remove population of the ground or excited states to any other far level and, thus, to determine in what state (excited or ground) is concentrated the information about photon echo in each period of time. Such experiments were not carried out yet.

Within the framework of concept about inequality of forward and reversed transitions in optics for an explanation of a nature of the photon echo it is not necessary to attract representations about a superposition of states. In each period of time the information about a photon echo is stored at one concrete level (ground or excited). Discussed in [2] illustration of a photon echo as an example of sports competition of runner actually is, obviously, absolutely exact physical analog of a photon echo process. The run on a circle corresponds to rotation or precession of atoms and molecules. The role of ”Maxwell’s demon”
can and should carry out the spin of photon. The absorption or emission of photon is accompanied by change of a rotational state of atom or molecule.

The photon echo can be considered as multiple stage process of four photon mixing, which includes stages of dephasing and rephasing of rotational motion. The corresponding scheme of energy levels is given on Fig.4. Here we shall consider common variant, when the longitudinal magnetic field is applied and the magnetic levels are not degenerated. Under action of radiation of the first laser pulse (P1) the absorption of photon and forward transition in the excited state takes place. The dephasing process, which is connected with inhomogeneity of rotational motion [33], here begins. Under action of the second laser pulse (P2) of the same frequency and polarization the stimulated emission of photon occurs and the atom comes back in the ground state. For a part of atoms, which had the appropriate orientation in space, this optical transition will be reversed. Its will return to an initial state and their "memory" will be "deleted". For other part of atoms such transition will be a forward one and the "memory" about the initial state will be kept. The rotation in the ground state for discussed case is absent and the dephasing process stops. Under action of radiation of the third laser pulse (P3) of other frequency and helicity a photon absorption and excitation of atoms in other excited state takes place, where the rotation occurs in the opposite side. The same heterogeneity of rotational motion results that the rephasing process begins. At time equal $t_{21}$ the rephasing process is finished and due to high cross-section of the reversed optical transition into the initial state the superradiation of a photon echo appears.

According to the given physical explanation the information about the photon echo after the first pulse is stored in the first excited state. After the second laser pulse (for a case of stimulated photon echo) it is only in the ground state. At last after the third pulse this information is concentrated in the other excited state. For usual two-pulse photon echo, when the stage between the second and third pulses is absent, the information about the photon echo during all period of its formation is stored only in the excited states. In this case under action of the second laser pulse two consistently optical transitions take place.

The scheme of Fig.4 was realized in work [34], where the shape of a stimulated photon echo in ytterbium vapor was experimentally studied. However later this scheme was reconsidered. In work [35] for an explanation of the same experimental results other experimental scheme was proposed. In this case the second pulse P2 has frequency different from frequency of the first pulse P1, and the frequency of the third pulse P3, on the contrary, coincides with frequency of the first laser pulse. The latter scheme, certainly, better corresponds to the concepts of coherency and superposition of states, but in this case the four-photon mixing is impossible. Therefore we believe, that the experimental scheme described in [34], is correct in contrast to the scheme, described in [35].

These interesting experiments in any case require continuation. From the text of [34] it is not clear, why the authors used this complicated scheme of photon echo. What will happen, if the frequency and helicity of the third laser pulse will be the same, as for the first two pulses:

1) the signal of photon echo will be the same,
2) it will be weak, 
3) it will be absent?

The phenomenon of a photon echo is one of many nonlinear optical effects, where the principle of inequality of optical transitions allows to understand physical sense of proceeding processes and to plan ways of their further experimental study.

4 Conclusion.

Thus, we have quite sufficient experimental proofs of inequality of forward and reversed transitions in optics. The major task here for the experimenters is to learn to measure the parameters of the reversed optical transitions. Before the theorists there is a problem of creation the mathematical model (alternative to the Bloch equations) for description the dynamics of optical transitions. Such model should include a principle of inequality of optical transitions [36]. The possible base of such model may be the Dirac equation, which in usual variant assumes time invariance violation in electromagnetic interactions [37]. The description of the physical phenomena with such model will have much more direct and clear physical sense, than the existing description.

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Fig. 1 Profile of absorption line of $SF_6$ molecules for the $\nu_3$ band $1 \leftarrow 0$ transition.

Fig. 2 Spectral dependence of the room temperature absorption cross-section of $SiF_4$ molecules. 1- spectrophotometer result. 2- Lorentzian profile with FWHM = 4.5 cm$^{-1}$.

Fig. 3 Spectral dependence of the room temperature absorption cross-section of $SF_6$ molecules. 1- spectrophotometer result. 2- Lorentzian profile with FWHM = 4.5 cm$^{-1}$.

Fig. 4 The basic scheme of a photon echo process.
Fig. 1 Profile of absorption line of SF$_6$ molecules for $\nu_3$ band 1---0 transition.
Fig. 2 Spectral dependence of the room temperature absorption cross-section of SiF₄ molecules.
Fig. 3 Spectral dependence of the room temperature absorption cross-section of SF₆ molecules.
Fig. 4 The basic scheme of a photon echo process.