Time reversal noninvariance in quantum mechanics and in nonlinear optics.

V.A.Kuz’menko
Troitsk Institute for Innovation and Fusion Research, Troitsk, Moscow region, 142190, Russian Federation.

Abstract
The experimental proofs of strong time invariance violation in optics are discussed. Time noninvariance is the only real physical base for explanation the origin of the most phenomena in nonlinear optics. The experimental study of forward and reversed transitions in oriented in uniform electric field molecules is proposed.

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Introduction
The idea of the arrow of time is quite natural and recognized in biology, chemistry and other fields [1]. However, its recognition in the physics unexpectedly turns out to be rather problematical because of the dynamical equations of the basic laws of physics are time reversal invariant. In classical theories the arrow of time is introduced as a result of time asymmetric boundary conditions with time symmetric dynamical equations [2, 3]. But for the full recognition of this concept the introduction of time asymmetry into the dynamical equations is a more preferable way [1]. And the nature gives us the grounds for such idea. However, this process, probably, will be rather long and hard [4, 5].

From our point of view the mistake of physicists is based on the two myths. The first myth is the opinion about absence of any experimental evidences of time invariance violation in electromagnetic interactions [1, 3, 6]. Below we shall discuss the three known for present day direct and independent experimental proofs of strong time invariance violation in optics. The second myth is the supposition about the existence of the so-called quantum coherent states [7]. This concept has now extremely wide spreading in physics. However, it turned out, that the concept of coherent states does not have any real physical base [8].

First myth
The interaction of polarized laser radiation with the specific non-magnetic metallic planar chiral nanostructures was studied in [9]. The authors believe that these experimental results unambiguously show the evidence of broken time reversal symmetry in such unusual object.

In other experiments the splitting and mixing of photons were studied [10, 11]. On the first stage the narrowband (∼ 0.01nm) radiation of nanosecond laser was transformed through down-conversion in the nonlinear crystal into two intense broadband beams (each spectral width ∼ 100nm). On the second

1Electronic address: kuzmenko@triniti.ru
stage this two broadband beams were mixed in the sum frequency generator [10] or in the process of two photon excitation of rubidium atoms [11]. The reversed process in these cases corresponds to mixing of the so-called entangled photons and leads to regeneration of the initial narrowband radiation. In contrast, the mixing of non-entangled photons should give broadband radiation and is the example of again only the forward process. Both experiments show the same results: the efficiency of reversed process is much greater, than the efficiency of forward process.

In the next case the forward and reversed transitions in $SF_6$ molecules were studied [12]. In those experiments the authors deal with such specific object as the so-called wide component of line in a spectrum of polyatomic molecules [13]. The forward transition ($CO_2$- laser photon absorption process) in this case has extremely high spectral width ($\sim 150$ GHz) and relatively small cross-section ($\sim 10^{-19}$ cm$^2$). In contrast, the reversed into the initial state transition (stimulated emission) has very small spectral width ($\sim 450$ kHz). The difference in spectral widths of forward and reversed transitions exceeds five orders of magnitude. Accordingly, the cross-section of the reversed process turns out to be in several orders of magnitude greater, than the cross-section of forward process.

So, for the present day we have quite sufficient quantity of direct experimental proofs of a strong time invariance violation in optics. We have also enormous quantity of indirect experimental evidences, which are connected with the second myth about the existence of coherent states.

Second myth

The concept of coherent states appeared and became all pervasive in quantum optics during last several decades. The coherent states usually are interpreted as specific states of atoms or molecules after its interaction with a coherent laser radiation. Most phenomena in nonlinear optics are usually explained as a result of interference of the coherent states [15, 16].

However, the close theoretical analysis of this concept in [8] has shown that the inability to measure the absolute phase of an electromagnetic field prohibits the existence of quantum coherent states [7, 17]. It deprives the concept of coherent states any real physical sense. The concept of coherent states is only a "convenient fiction" for physicists in the field of quantum optics [8].

Physical origin

We believe that the only real physical base of observed phenomena in nonlinear optics is the time reversal noninvariance in electromagnetic interactions or inequality of forward and reversed processes in the optics. So, most phenomena, which are explained now as the interference of the coherent states, are really a manifestation of the time reversal noninvariance. However, time noninvariance manifests itself usually only in indirect way and this is the main reason why the concept of coherent states is popular till now.

What is the usual origin of inequality of forward and reversed processes in optics? The discussed above experiments show extremely high efficiency of the reversed process. However, for experiments with splitting and mixing of photons
the origin of this efficiency is not clear as a whole. It is connected with the subtle concept of entanglement and it is the problem for future studies.

For optical transitions in atoms and molecules the high efficiency of the reversed process is unambiguously connected with its high cross-section. However, we do not need to have a doubt in equality of the Einstein’s coefficients for forward and reversed transitions [18]. The equality of Einstein’s coefficients means only the equality of integral cross-sections of the opposite processes and does not prohibit the inequality of differential cross-sections. Such possibility is well illustrated by the discussed above experiments with SF₆ molecules [12]. In this case the high cross-section of reversed optical transition is connected with its extremely small spectral width in contrast to the forward transition.

The wide component of line is rather specific object and it exists only in the large polyatomic molecules. In small molecules and in atoms the line wings of such kind are absent. What is the reason in these cases of high differential cross-section of the reversed optical transition?

Here we should pay attention to the quantum mechanical averaging process of some parameters of vibrational and rotational motions, which manifests itself in a molecule absorption spectrum. The lines in absorption spectrum of small molecules are very narrow. Its frequency allows calculating the moment of inertia of molecule with precision \( \sim 0.0001\% \). From other side the temporal change of moment of inertia during the period of vibrational motion of the atoms in molecule usually exceeds the value of 1% [19]. This inevitably means that there exists some quantum mechanical process of averaging of moment of inertia during the period of vibration of the atoms in molecule. It is worth to mention that for polyatomic molecules this process of averaging, obviously, can undergo some short reversible violations even for the forward transition, which lead to appearance in absorption spectrum of the so-called line clumps [20].

The time asymmetry may consist in the difference between the averaging processes for forward and reversed transitions. For the forward transition the absorption cross-section does not depend from the phase of vibration motion. In contrast, for the reversed transition such dependence may exist. If the molecule has the phase of vibration motion, which allows it to return exactly into the initial state, then the backward transition will be reversed and its differential cross-section will be much higher than the averaged cross-section of the forward transition. In other cases the differential cross-section of the backward transition will be relatively small and such transition should be called over again only as the forward one. Inequality of forward and reversed processes in a natural way supposes the existence of the memory of atoms and molecules about the initial state. In some sense such memory can correspond to the entropy of quantum system.

So, the discussed above supposition is a good physical base for explanation of the origin of experimental study of the dynamics of vibration motion of atoms in molecule [21].

**Oriented molecules**

The similar quantum mechanical process of averaging of cross-section, prob-
ably, takes place for the rotational motion also. The rather common opinion exists, that the cross-section of interaction between the molecules and radiation should depend from orientation of molecules with respect to laser beam [22]. Some experiments with the anisotropy of fluorescence or with the successive transitions confirm this supposition. However, as a whole this is rather knotty problem. Practically all experiments in this field are carried out only with the linearly polarized laser radiation. Some authors even talk about the absorption of a linearly polarized photon [23]. Such photon, of course, is absent in nature. The linearly polarized light is a complex object and consists of the equal quantity of photons with different spins. So, we believe, that for the first stage of experiments with oriented molecules the more simple circularly polarized light should be preferably used.

The process of orientation averaging of absorption cross-section is well illustrated, for example, by the experimental results of work [24], where the absorption of laser radiation by the hydrogen cyanide trimer molecules in the static electric field was studied. The external static electric field interacts with the dipole moment of molecule and tries to orientate it in the space. The experiments show, that when the energy of this interaction is smaller, than the energy of rotation motion, the perturbation of rotation motion practically does not manifest itself in the absorption spectrum. In the opposite case the molecules turn into the so-called pendular states and the absorption spectrum dramatically changes.

The inequality of forward and reversed transitions, again, may be the result of different character of quantum mechanical processes of orientational averaging of cross-section of molecule interaction with laser field. For simplified illustration the Fig.1 shows the assumed dependence of transition’s cross-section from the angle between the molecule axis and the direction of laser beam for forward (1) and reversed (2) transitions. The integral cross-sections of both transitions are equal. If the molecule is oriented in the space so that the backward process returns it exactly into the initial state, then the differential cross-section will be very large and such transition is reversed. In other orientation of molecule the backward process has relatively small differential cross-section and it should be called again only as the forward transition. In this case the molecule remains the memory about its initial state. So, in such way the origin of the so-called rotational coherency may be explained [25].

The other indirect evidence of inequality of forward and reversed transitions is the numerous experiments with degenerated or nondegenerated four photon mixing in the so-called folded boxcars arrangement [21]. Here the three laser beams of different directions are crossed in common point in a liquid, gas or in a molecular beam. The appearance of the superfluorescence in the new direction is observed. The cross-section of transition for such superfluorescence should be extraordinarily high. The direction and temporal characteristics of appearance of superfluorescence correspond to such orientation and phase of vibrational motion, which allow molecules to return exactly into the initial states.

New and detailed information about the orientational dependence of cross-sections of forward and reversed transitions may, probably, be obtained in the
experiments on the existent apparatus, which uses the static electric fields and molecular beam with cryogenic bolometer [12, 24, 26]. The Fig.2 shows the simplified arrangement for suggested experiments. The radiation of infrared laser is split on the two beams (pump and probe). The interaction regions are placed between the flat electrodes, which have slits for laser radiation and can be independently rotated round the molecular beam. As the object for experimental study the stable linear molecules with large dipole moment and small rotational constant well suit. There are, for example, HCCCN or H(CC)nX molecules, were n=1–3 and X - is the halogen. The low rotational states of such molecules may be easily turned into the pendular states at a relatively weak electric field. For the reversed optical transition we can expect, that the experimental dependence of its cross-section from the angle between the pairs of electrodes will be rather similar to the dependence on Fig.5 in work [12]. As a whole, such experiments can give important information about the orientational dependence of cross-sections for forward and reversed optical transitions.

Conclusion

Time reversal noninvariance is the only real physical base for explanation the origin of the most phenomena in nonlinear optics. For present day we have quite sufficient quantity of the direct experimental proofs of the strong time invariance violation in optics. This is a good reason to introduce the time asymmetry into the dynamical equations of the basic laws of physics. The corresponding asymmetric equations for description the dynamics of optical transitions will substitute for the famous Bloch equations, which are widely and successfully used in optics now [15], but which does not have any clear physical sense. The experiments for the study of orientational inequality of forward and reversed transitions in molecules are proposed.

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Fig. 1  The supposed shapes of dependences of cross-section from the angle between molecule axis and the direction of the laser beam for forward (1) and reversed (2) transitions.
Figure 2. A schematic diagram of the apparatus for proposed experiments with oriented molecules. Bs - beamsplitter, M - flat mirror, E1 - rotated electrodes with slits.