Femtosecond laser as a tool for experimental study of time invariance violation in optics.

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

Abstract
The experiments with weak collinear probe pulses for time reversal noninvariance study in optics are proposed and discussed.

PACS number: 42.50.Hz

The equivalence of forward and reversed processes is a fundamental concept in optics today. However, this is only the hypothesis which does not have any experimental proofs. In the nearest future this concept should be rejected. From one side, because of it gives very meager possibility to explain physical origin of great number of phenomena in nonlinear optics. Frequently such phenomena have good mathematical description on the base of Bloch equations. However, these equations do not have clear physical sense. The concept of interference of "wave packets" or coherent states of molecules is usually used for explanation of the origin of nonlinear phenomena. However, the concept of coherent states in itself does not have clear and reliable physical base [1,2]. The concept of inequality of forward and reversed processes is the alternative analog of the concept of coherent states. In contrast to latter concept, it has clear physical sense and gives good base for explanation of the origin of number of phenomena in nonlinear optics [3].

From other side, the equivalence concept should be rejected because of the opposite concept even today has several direct experimental proofs [4]. However, the concept of time noninvariance is so radical for optics, that it is not clear today how many experimental proofs will need for its recognition. A femtosecond lasers should play important role here.

The obtained experimental results show, that the difference in cross-sections of forward and reversed processes can exceed many orders of magnitude [5, 6]. The cross-section of reversed transition, obviously, has very sharp dependence from the orientation of molecules in space, from the phase of vibrational motion of atoms and even from the phase of laser radiation in the space. The classical pump-probe experiments with femtosecond laser pulses will give, probably, the most exact information of such kind. In such experiments two demands must be fulfilled:

1) - the pump and probe beams should be exactly collinear,

2) - the pump pulse should not saturate optical transition and the probe pulse should be very weak avoiding changing the distribution of species on the

[1]Electronic address: kuzmenko@triniti.ru
energy levels.
Only such arrangement allows to obtain correctly information about the cross-section of reversed optical transitions. The principle set up of such experiments is shown in Fig. 1a, where a fluorescence of excited species is used in a detection scheme.

It is surprising, but among the huge number of pump-probe experiments with femtosecond lasers such experimental arrangement is not used till now. The weak probe pulse is usually used in the cross beams geometry. This case suits for four photon mixing, but does not suit for the study of the reversed transitions into the initial state.

The most closely related experiments were carried out in [7-10]. The authors studied the fluorescence of molecules or atoms (which characterizes the population of excited states) after it interaction with the pair of collinear phase-locked femtosecond laser pulses. However, the authors used equal intensity of pump and probe pulses. Such experiments should be continued in low intensity regime with weak collinear probe pulse. Close to ideal intensity regime was used in [11], but the demand of collinearity was not fulfilled. In proper conditions some amplification of probe laser radiation without inversion should be observed in some degree similar to results of [5]. It allows to make the evaluation of difference in cross-sections of forward and reversed transitions.

Here, the widely spread mistake exists that femtosecond laser radiation can align molecules in the so-called field free regime [12, 13]. This point of view does not have reliable experimental proofs. Our explanation is that the observed revivals are the manifestation of very sharp dependence of the reversed Raman transition’s cross-section from orientation of molecule in the space [14]. The discussed low intensity collinear pump-probe experiments will make this situation clear. The principle scheme for reversed Raman transitions study is shown in Fig. 1b. As a detection tool, the high harmonic generation (HHG) [15], the Coulomb explosion imaging [16] or polarization [17] techniques may be used. The pump femtosecond pulse produces forward Raman transitions between the ground states of molecules. The strong reading pulse is fixed in time and turned on the strong delayed revival. The weak collinear probe laser pulse is used for study the efficiency of reversed Raman transitions. Rather close experiments were carried out recently in [17, 18]. However, again the intensity of probe pulses was too strong.

As a detection scheme in such experiments a four photon mixing phenomenon in the so-called boxcars arrangement may be used [19, 20]. In such way the high quality experimental result with Raman transitions were obtained recently in [21]. It demonstrates not the suppression of alignment, but the erasing of stored information by efficient reversed process under action of the probe pulse (as a whole quite similar to those in [10]). The additional experiments with low intensity collinear probe pulses will give some information about relative cross-section of the reversed Raman transitions into the initial state.

In conclusion, we discussed the experiments with weak collinear probe femtosecond laser pulses for time invariance violation study in optics. Such experiments will allow us to understand and to give clear physical explanation.
for number of nonlinear phenomena, like as the population transfer or so-called
"coherent control" [22].

References

[1] K.Mølmer, Phys.Rev.A 55, 3195 (1997).
[2] K.Nemoto, and S.L.Braunstein, E-print, quant-ph/0312108.
[3] V.A.Kuz'menko, E-print, physics/0306148.
[4] V.A.Kuz'menko, E-print, physics/0506023.
[5] C.Liedenbaum, S.Stolte, and J.Reuss, Chem.Phys. 122, 443 (1988).
[6] B.Dayan, A.Pe'er, A.A.Friesem and Y.Silberberg, E-print, quant-ph/0401088.
[7] N.F.Scherer, A.J.Ruggiero, M.Du, and G.R.Fleming, J.Chem.Phys. 93, 856 (1990).
[8] N.F.Scherer, R.J.Carlson, A.Matro, M.Du, A.J.Ruggiero, V.Romero-Rochin, J.A.Cina, G.R.Fleming, and S.A.Rice, J.Chem.Phys. 95, 1487 (1991).
[9] H.Yamada, K.Yokoyama, Y.Teranishi, A.Sugita, T.Shirai, M.Aoyama, Y.Akahane, N.Inoue, H.Ueda, K.Yamakawa, Y.Suzuki, M.Kawasaki, and H.Nakamura, Phys.Rev. A 72, 063404 (2005).
[10] K.Ohmori, H.Katsuki, H.Chiba, M.Honda, Y.Hagihara, K.Fujii, Y.Sato, and K.Ueda, Phys.Rev.Lett. 96, 093002 (2006).
[11] V.I.Prokhorenko, A.M.Nagy, and R.J.D.Miller, J.Chem.Phys. 122, 184502 (2005).
[12] E.Peronne, M.D.Poulsen, C.Z.Bisgaard, H.Stapelfeldt, and T.Seideman, Phys.Rev.Lett. 91, 043003 (2003).
[13] I.V.Litvinyuk, K.F.Lee, P.W.Dooley, D.M.Rayner, D.M.Villeneuve, and P.B.Corkum, Phys.Rev.Lett. 90, 233003 (2003).
[14] V.A.Kuz'menko, E-print, physics/0310090.
[15] K.Miyazaki, M.Kaku, G.Miyaji, A.Abdurrouf, and F.H.M.Faisal, Phys.Rev.Lett. 95, 243903 (2005).
[16] H.Stapelfeldt, and T.Seideman, Rev.Mod.Phys. 75, 543 (2003).
[17] M.Renard, E.Hertz, S.Guerin, H.R.Jauslin, B.Lavorel, and O.Faucher, Phys.Rev.A, 72, 025401 (2005).
[18] K.F.Lee, E.A.Shapiro, D.M.Villeneuve, and P.B.Corkum, Phys.Rev.A 73, 033403 (2006).

[19] Y.Prior, Appl.Opt. 19, 1741 (1980).

[20] M.Schmitt, G.Knopp, A.Materny, and W.Kiefer, Chem.Phys.Lett. 270, 9 (1997).

[21] Sh.Fleischer, I.Sh.Averbukh, and Y.Prior, E-print, quant-ph/0601197.

[22] M.Dantus, and V.V.Lozovoy, Chem.Rev. 104, 1813 (2004).
Fig 1  Principle experimental setup for pump-probe study of the reversed transitions: a) for excited states of molecules with fluorescence detection and b) for Raman transitions with HHG detection. D – detector, G – grating.