Femtosecond laser as a tool for experimental study of inequality of forward and reversed processes in optics.

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Abstract

The classical pump-probe scheme for experiments with femtosecond laser is proposed to use for study the inequality of forward and reversed transitions in optics.

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Rather usual situation in nonlinear optics is a case, when the mathematical description of phenomenon is not accompanied by corresponding clear explanation of physical nature of this phenomenon. The most significant example is the effect of adiabatic population transfer in a two level system after sweeping of resonance conditions. The mathematical model of rotating wave approximation gives excellent description of phenomenon, but its physical explanation is absent [1,2].

Similar situation exists in the field of rotational coherence spectroscopy [3]. Corresponding mathematical model well describes the temporal characteristics and the shape of observed rotational transients (revivals) [4]. As a physical explanation of the nature of transients the concept of quantum interference of coherent states is usually used [3]. However, the opinion exists that the concept of coherent states has a not reliable physical base [5-7]. A good alternative here is the concept of inequality of forward and reversed processes in optics [8]. Now we have not only indirect evidences, but also the quite direct experimental proofs of such inequality [9-11].

The experiments show that the reversed process can have extremely high efficiency. For optical transitions it corresponds to very high cross-section of reversed transition into the initial state. However, the integral cross-sections of forward and reversed transitions (Einstein’s coefficients) should be equal. Some indirect evidences allow to suppose the following shape of orientation dependence of cross-section for reversed transition: extremely high cross-section for transition into the initial state and relatively small cross-section for other orientation of molecules in space (Fig.1). In this case the explanation of physical origin of rotational coherence and population transfer effects become quite simple and clear.

The main way for experimental study of transition cross-section dependence from orientation of molecule relative to the direction of laser beam is quite clear: we can use a sequential optical transitions [12]. The radiation of the first laser...
selects the molecules from isotropic distribution, the second laser beam deals
with the aligned molecules and the third laser beam will deals with the oriented
molecules. However, any complete and extensive experimental works of such
kind are absent in literature.

Some negative role here, obviously, plays the concept of so-called pendular
states [12]. It is supposed, that intense laser radiation can break a free rotation of
molecules and align it about the laser polarization axis. Extensive experimental
works in this field, however, can not give any reliable proofs, that this effect
really exists [12]. The alternative explanation of the experimental results, which
includes the concept of inequality of forward and reversed transitions, is quite
possible [13].

Pump-probe experiments with femtosecond laser may be used for study the
orientation spectra of both forward and reversed transitions. Large number of
pump-probe experiments in the field of rotational coherence spectroscopy are
described in literature. But in most cases the energy of the probe pulse is equal
to the energy of the pump pulse [4,14,15] or even exceeds it [16]. The scientists
usually take an interest in temporal characteristics and in shape of revivals.

The main purpose of this note is to propose to use for experimental study
of cross-sections of forward and reversed transitions the classical pump-probe
scheme, when the energy of probe pulse is much smaller, than the energy of
pump pulse. In this case the radiation of probe pulse does not change substan-
tially the distribution of molecules after the pump pulse and does not make
difficulties for interpretation of experimental results. Measured amplitude of
transients contains information about the cross-section of forward and reversed
transitions.

Two different proposed experimental schemes are presented in Fig.2. In the
first case (Fig.2a, where the excited state takes part) the dependence of integral
fluorescence intensity of molecules is studied from the delay time between pump
and probe pulses. The energy of the probe pulse should be 0,01 - 0,05 of the
pump pulse. It is advisable to modulate intensity of probe beam and to use
a phase-sensitive detection scheme for precise measurement of absorption or
amplification of probe pulse radiation. We can expect to observe very strong
transients with even some amplification of probe pulse (similar in shape as
in Fig.5 of Ref. [9]). If we know the fraction of molecules excited by the
pump pulse, we can determine the ratio of cross-section of forward and reversed
transitions.

The second case is very similar to work [4] (Fig.2b, Raman variant without
an excited states of molecules), where the modification of polarization of reading
pulse is measured. This reading pulse is tuned to one of the far transients. The
probe pulse has the same polarization as the pump pulse. In this case it is also
helpful to use the phase-sensitive detection scheme with modulated probe beam.
We can expect, that the scanning of the probe pulse will give some decreasing
of measured signal due to efficient transfer of molecules into the initial state.
The dependence of signal from intensity of pump and probe pulses will give
information about the cross-section of forward and reversed transitions.

In conclusion, we expect that the discussed experiments will make clear a
physical origin of large number of phenomena in nonlinear optics.

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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.
Fig 2. Proposed experimental setup for pump-probe study of the reversed transitions: a) for the case with excited states of molecules and b) for the Raman case without excited states of molecules. PM- photomultiplier, P- polarizer, A- analyzer.