

Comment on “Long-lasting field-free alignment of large molecules inside helium nanodroplets”

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A physical explanation for the effect of suppression of ionization of molecules upon overlapping of a long low-intensity alignment pulse and a strong short probe pulse is proposed.

Experiments on the so-called alignment of large molecules in helium nanodroplets are described in [1]. The idea of aligning molecules in the field of intense nonresonant laser radiation has been popular for many years [2]. It is supposed, that the dipole moment induced in a polarizable molecule by the laser field can hinder free rotation of molecules and align molecules along the laser polarization vector. This old myth has no reliable experimental proofs. In reality, we are dealing with a combination of the so-called geometric alignment (when the probability of absorption of a photon depends on the orientation of the molecule in space [3, 4]) and the inequality of the differential cross sections for forward and reversed processes in quantum physics [5].

However, the subject of this comment is different. In [1] the interesting experimental fact was found: during a long alignment pulse, ionization of molecules by an intense short probe pulse is substantially suppressed. Ionization becomes effective only after a sharp interruption of the alignment pulse (Fig. 3). The explanation of the authors is that the radiation of the alignment pulse causes fragmentation of the detected molecular ions.

It is obvious that this is a wrong explanation. The radiation of the alignment and probe pulses has the same wavelength. At the same time, the radiation intensity of the alignment pulse is hundreds of times smaller than the intensity of the probe radiation. It can't significantly affect the formed molecular ions. The experiment also shows that the effect of radiation dose accumulation is absent.

So, here we are dealing with a variant of the widely known phenomenon - coherent population trapping. This is one more old myth. It is assumed that quantum interference modifies the optical properties of the quantum system in such a way, that the so-called dark states appear [6]. In the dark state the atoms or molecules neither absorb nor emit a light. However, the matter isn't that atoms or molecules don't absorb radiation, but that they radiate it very well when they transfer to the initial state [7].

We believe that the physical nature of the discussed effect is explained by the competition of two processes: the direct process of excitation and ionization of molecules and the reversed process of deactivation of molecules due to stimulated emission, which transfers molecules to the initial quantum state. In Fig. 1 the scheme of the proceeding processes is submitted. Under the action of the radiation of the alignment pulse, a direct process of Raman transitions between degenerate quantum levels of molecules occurs. It has a small differential

cross section. In this case, the quantum system in the state (b) has a memory of the initial quantum state (a). Further we have the competition of two processes:

- 1) Absorption of several photons from the radiation of the probe pulse and ionization of the molecule. Moreover, the absorption of the first photon is a partially reversed process and has a larger differential cross section than the absorption of subsequent photons.
- 2) At the same time, the radiation of a powerful probe pulse facilitates the stimulated emission of photons and the return of molecules to the original quantum state (a) [8]. This process of stimulated emission is the closest to fully reversed and has a very large differential cross section. It lies at the heart of such a long-known effect as electromagnetically induced transparency [8, 9].

In the case of overlapping in time alignment and probe pulses, the stimulated emission has an advantage and the ionization of the molecules turns out to be suppressed. In the absence of the radiation of the alignment pulse, the direct process of photon absorption from the probe pulse and the subsequent ionization of the molecules gain an advantage.

We can expect that such effect should also be observed in high harmonic generation experiments [10].

Thus, we need an experimental investigation of the differential cross sections of forward, reversed and partially reversed quantum processes. These differential cross sections are a direct consequence of the fundamental property of quantum physics – its time reversal noninvariance [7].

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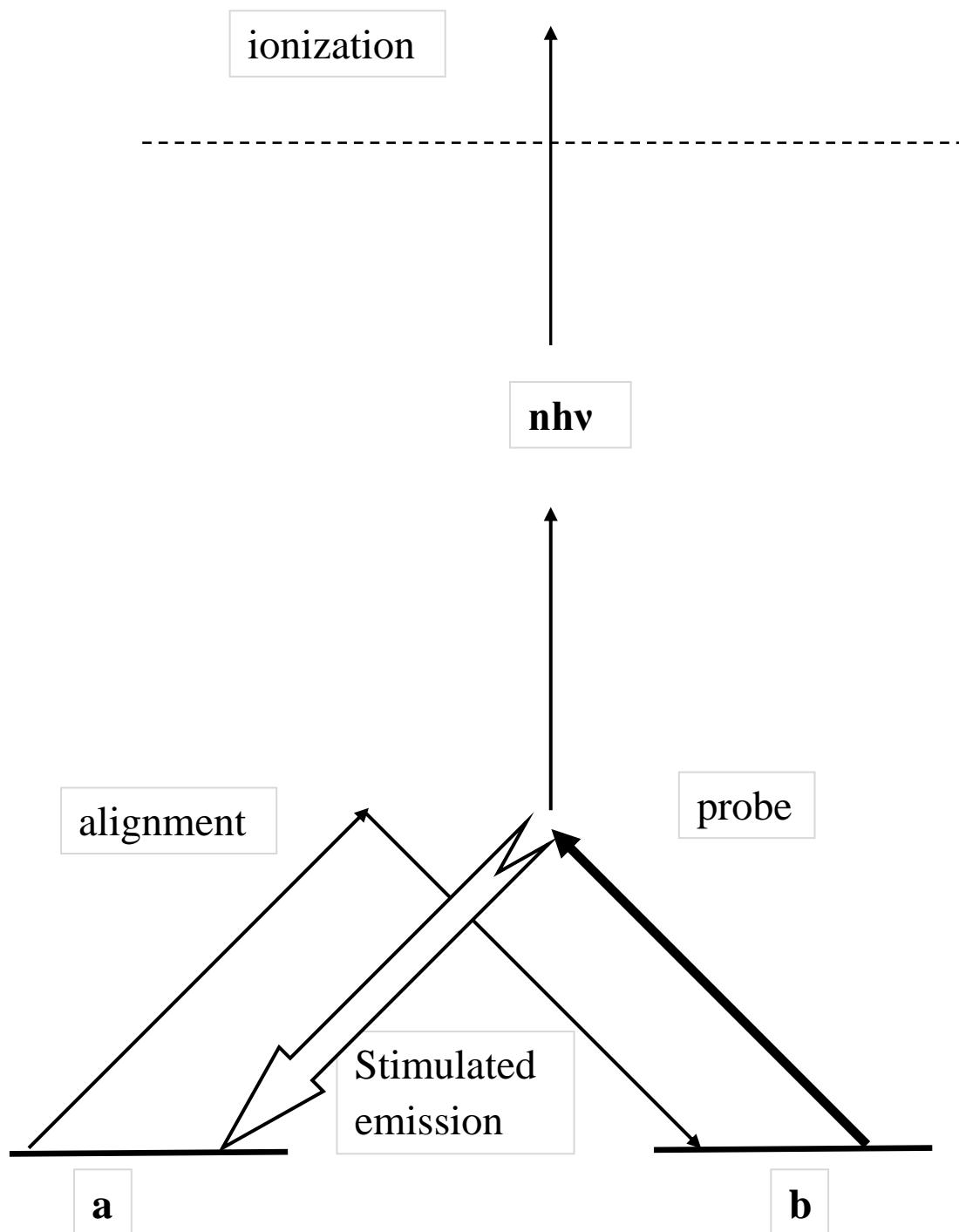


Fig. 1 The scheme of processes, demonstrating the competition of the reversed process of stimulated emission and the direct process of photon absorption and ionization of molecules.