Temporal Coherent Control on Multiphoton Transitions Using Incoherent Light

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An interferometric technique involving two delayed short laser pulses in resonance with a energy transition of a quantum system (time delay spectroscopy, TDS) has been successfully used for extracting spectral information in atomic and molecular systems as well as in semiconductors[1, 7]. In its simplest form a short pulse induces coherence between two levels of the system, thus creating a polarisation in the medium. The induced polarisation oscillates at the transition frequency with a decreasing amplitude during the dephasing time. The second pulse, depending on the phase of the polarisation oscillation when starts its interaction with the medium, can enhance or destroy the residual polarisation and the population of the upper level. As a result, the population at the end of the second pulse exhibits interference fringes versus the delay between the two pulses. This effect, due to the quantum memory of the atomic system, is known in literature as quantum interference[8]. Information on the energy differences between nearby states in the atom and the dephasing time of the polarisation, i. e. the line shape of the transition, are extracted from the time modulation of the fringe pattern[4]; the spectral resolution is given here by the maximum value of the delay. The technique has been applied to both single and multiphoton transitions[8]. In the latter case, it has been shown that, in opposition to the one-photon case, one can distinguish between optical interference, which occurs when the two pulses overlap temporally, and quantum interference, which takes place even when the two pulses are well separated in time.

Nearly all the TDS experiments reported in literature have been performed by using transform-limited laser pulses, with duration shorter than the characteristic times of the system. However, it is not necessary to use excitation pulses of such characteristics in order to observe a quantum interference effect. For example, in a one photon transition in weak-field regime, it is easy to demonstrate that two equal non transform-limited pulses give rise to a quantum interference pattern even when their duration is much longer than the system correlation time. An experimental evidence of such a principle has been given in ref.[9], by observing the population of a manifold of Rydberg states excited by one-photon absorption.

In this contribution we investigate the equivalence between the cases of transform-limited and not transform-limited laser pulses in a resonant multiphoton ionisation process. We study

a three-photon two-photon resonant ionisation of atomic barium performed with a sequence of two identical, not transform-limited, laser pulses. We show that the experimental results, obtained with two pulses of a multimode laser with 27 GHz spectral bandwidth and 5 ns duration, can be reproduced theoretically using two transform-limited pulses of 33 ps duration having the same spectral bandwidth of the experimental ones. In conclusion we show that nanosecond not trasform limited pulses can be used for the time delay spectroscopy technique even for multiphoton processes instead of shorter time duration sources. An imediate advantage is the much easier tunability of the nanosecond sources with consequent great range of applications.

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