Two-photon coherent control of femtosecond photoassociation

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Photoassociation (PA) has emerged as a technique to create ultracold ground state molecules. The difficulty lies in the unfavorable Frank Condon overlap between the wavefunctions of the colliding atom pair and the molecular ground state. Therefore such a scheme requires a series of excitation and deexcitation steps. A broad band excitation containing all these transitions seems a natural choice [1]. In order to discriminate between the desired goal of reaching the ground state and all other possible outcomes, coherent control schemes have to be adopted to PA. However, one of the main obstacles is to eliminate the atomic excitation which hampered first attempts at femtosecond PA [2]. Indeed, the broad bandwidth of femtosecond lasers appears to be an obstacle for PA rather than a tool for control. A possible remedy is to reduce the bandwidth from femtosecond to picosecond pulses [3]. But picosecond pulse shaping has yet to be developed and to date the reduced bandwidth limits the possibilities of control.

To overcome the ostensible conflict of driving a narrow transition by a broad-bandwidth laser, we explore a two-photon photoassocation scheme. An antisymmetric phase pattern [4] can be employed to eliminate the atomic line. This is based on the fact that excitations carried out by more than one photon can take advantage of constructive and destructive interferences which lead to different pathways of the field. In addition further control of molecular pathways can be achieved. While two-photon absorption may pave the way toward coherent control of PA, the success of two-photon coherent PA will ultimately depend on the pair density of atoms at short enough internuclear distances where PA is efficient. The possibility to enhance the pair density via control of scattering resonances will be discussed.

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