

Directional Quantum Controlled Chemistry: Generating Alignment of Ultracold Photoassociation Molecules

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Quantum coherent control was conceived as a method to direct chemical reactions to a desirable outcome [1],[2],[3]. Despite the remarkable success of the application of control methods to induce uni-molecular manipulations like photo-isomerization[4], photo dissociation[5], the raison d'être for coherent control in the context of chemistry, namely, the assembling of multiple atoms or molecules into a new molecule, was not realized until recently [6],[7]. The main obstacle has been the pre-entanglement of the reactants [8]. The approach to overcome this issue differs in hot and cold collisions. For reactions in the high ($T \gg 1K$) temperature regime, one has to first distill entangled pairs out of thermal incoherent mixture [9]. In the cold ($T \ll 1K$) regime, quantum phenomena close to threshold effects the low angular momentum states are pre-entangled. For the ultracold reactions, technological developments that gave access to the instant frequency in the time domain [10] were used to demonstrate coherent control effects for ultracold photoassociation[7].

We present a new aspect of quantum control of chemical reactions unique to photochemical reactions. Such control is able to align the products of the reaction along a well-defined spatial direction relative to the lab frame. We show that under conventional shaped photo-associated pulses in the ns regime a significant alignment is developed. We explore the dependency of and give several detecting approach

A theoretical investigating in pulsed photoassociation of ultracold ^{87}Rb atoms shows significant alignment of a specific vibrational state $v'' = 39$ on the ground $^3\Sigma_u^+$ electronic state. The simulation is based on solving the time dependent Schrödinger equation with the pulse contained explicitly and performing thermal averaging. Significant alignment is obtained by transform limited pulses. Enhancement of the population transfer by chirped pulses slightly decreases the alignment. Employing multipulses in synchronization with the vibrational and rotational periods leads to coherent enhancement of both population and alignment of the target state.

A linearly polarized REMPI probe that will be tuned to an electronic Π state induce electronic excitations perpendicular to the molecule axis. For molecules which are aligned to the lab fixed axis a difference in the molecular signal for REMPI pulse polarizations parallel and perpendicular to the pump should give a clear indication for the alignment. For the values that presented in this paper one can estimate the ratio of the signal to be at most at the order of $S_{\perp}/S_{\parallel} \sim 3$. Once the photoassociation generates a $v'' = 39$ molecule, additional alignment can be obtained by excitation de-excitation cycles leading to lower vibrational states.

The introduction of lab-frame spectroscopy into the coherently controlled ultracold reaction could raise several interesting applications. For heteronuclear ultracold molecules that possess permanent dipole moment, a controlled photoassociation can generate macroscopic *orientation* of whole ensemble. Furthermore, under BEC conditions, the induced macroscopic dipole could exhibit phenomena that originate in the quantum characters of the molecules but give signatures on the macroscopic scales. A concerted rotation of the quantum condensed phase can lead to effects which will have analogs in other classical-like condensed phase dynamics.

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