Femtosecond Laser Control of the Chemical Reaction of Carbon Monoxide and Hydrogen

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Abstract: Femtosecond laser control of chemical reactions is made possible through the use of pulse shaping techniques coupled to a learning algorithm feedback loop – teaching the laser pulse to control the chemical reaction. This can result in controllable relative fragmentation ratios for unimolecular dissociation reactions – therefore providing selectivity in breaking of bonds in a molecule. More interestingly, the same techniques can be used to provide control over chemical reactions involving two or more reactant molecules, where these come together, react and produce different reaction products. Specific high-value reaction products can possibly be produced selectively, without need for further separation or purification. In this work, we focus on low pressure gas phase laser induced chemical reactions, with the aim of controlling the reaction between CO and H_2 to produce hydrocarbon products, of interest to the petrochemicals industry. Preliminary experimental results will be presented.

1. Introduction

Coherent control of chemical reactions is a major goal in chemical physics and much progress has been made in this field over the last 15 years, see reviews [1-3]. In particular, coherent control of molecular dissociation in low pressure molecular beams has been demonstrated by various groups, the first example of selective bond breaking was demonstrated by Assion et al [4]. In addition, some work has been done at higher pressures and in liquid phase [5,6]. Practical application of coherent control techniques requires work to be done on liquid phase and under high pressure gas phase conditions.

2. Results

Preliminary experiments were conducted with transform limited 130 fs, 1 mJ laser pulses at 795 nm in an effusive molecular beam at an average chamber pressure of 5 x 10^{-5} mbar. The reaction probability is expected to be much lower due to much less collisions occurring under these conditions, but we nevertheless find the interesting and exciting result of peaks at masses 29 and 30, which would correspond to CHO and CH₂O, two potential intermediate products in the Fischer-Tropsch reaction [7]. More work will be presented in further validation of these and other potential intermediate species and potential control of reaction products in this reaction.



Figure 1: Mass spectrum of a 1:1 gas mixture of CO and H_2 in a hot molecular beam, after irradiation with 130 fs, 1 mJ laser pulses. There is some evidence for the formation of CHO and CH₂O, expected intermediate products in the Fischer-Tropsch reaction.

3. References

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