



FRONTIERS IN OPTICAL COHERENT AND ULTRAFAST SCIENCE

A NATIONAL SCIENCE FOUNDATION PHYSICS FRONTIER CENTER AT
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A nugget from FOCUS:

Control of nonlinear resonant photochemistry of 1,3-cyclohexadiene in solution

E. C. Carroll, B. J. Pearson, A. C. Florean, P. H. Bucksbaum, and Roseanne J. Sension

The photochemistry of small organic molecules such as cyclohexadiene (C_6H_8) is an important interface of physics, chemistry and biology. Often these reactions happen within a few hundred femtoseconds. On this ultrafast timescale, the dynamics of the reaction can be controlled by changing the “shape” of the laser pulse. Discovering which properties of the laser pulse facilitate or inhibit a reaction is a powerful tool for understanding the quantum mechanics behind photochemical reactions.

We have used learning algorithms and pulse shaping techniques to study the ring-opening reaction in cyclohexadiene (CHD). When CHD is electronically excited, 4 times in 10 it will rearrange into the linear molecule 1,3,5-hexatriene. A similar process occurs in human skin in the photochemical formation of Vitamin D.

In this experiment, the CHD molecule must absorb three photons before it can form hexatriene. This kind of multi-photon absorption typically occurs best when the laser pulse is very intense, as the probability of three photons being incident on a CHD molecule simultaneously is highest.

Surprisingly, however, in the multi-photon CHD reaction, the most intense laser pulses do not lead to the most hexatriene. The learning algorithm discovered that it is important for the high energy components of the laser pulse to precede the lower energy components. This property of the laser pulse (negative chirp) reduces the chance that a molecule will become excited, but *increases* the probability that if it is excited, it will undergo the reaction.

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