STUDY OF VINYLIDENE-ACETYLENE ISOMERIZATION BY COULOMB EXPLOSION IMAGING

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C$_2$H$_2$ is one of the simplest molecules that exhibit intramolecular rearrangement in a vibrationally excited state. The nature of the vinylidene-acetylene isomerization mechanism has been the focus of much theoretical investigation. The calculations predicted the existence of a vinylidene (C=CH$_2$) isomer 2 eV above the acetylene vibrational ground state. Experimentally, the vinylidene has been studied by photoelectron spectroscopy · a revealing resonances which have been assigned as vibrational vinylidene structure. The neutral vinylidene has been predicted to decay into linear acetylene configurations due to the low isomerization barrier of < 0.2 eV. This decay time has been estimated to be 0.2-4 psec · b.

We applied the Coulomb explosion imaging (CEI) technique to measure the structure of C$_2$H$_2$ molecules which were produced by electron photodetachment from vinylidene C$_2$H$_2^-$ anions · c. The nuclear density of C$_2$H$_2$ molecules as measured by CEI 3.5 µsec after their production as vinylidene isomers shows unambiguously that a large fraction (∼50%) of the molecules retain the vinylidene conformation. This surprising result contradicts the accepted concept of a rapidly “decaying” vinylidene isomer.

A further analysis of the CEI results suggests a different interpretation of the isomerization process. The number of total molecular eigenstates with vinylidene character is estimated to be only $N = 2$ - 4. Therefore, on a time scale shorter than the characteristic time of radiative decay, no “decay” of vinylidene into acetylenic configurations occurs. The structure of these excited states is better described as a coherent superposition of both acetylene and vinylidene amplitudes.

In this presentation, a detailed analysis of the measured structure and comparison with recent theoretical calculations will be given.

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