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Alignment of H_2 (E,F) under intense laser fields

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Abstract

Velocity map imaging (VMI) is used to study the photophysics of the H_2 during 532 nm non-resonant photodissociation, following excitation to (E,F) state prepared by resonance enhanced multiphoton ionization (REMPI).
Photoelectrons and photofragment images have been recorded through the 5 MW/cm² to 160 TW/cm² power range. Alignment with an external field source (1064 nm) during the dissociation has been probed from 50 MW/cm² to 500 MW/cm².

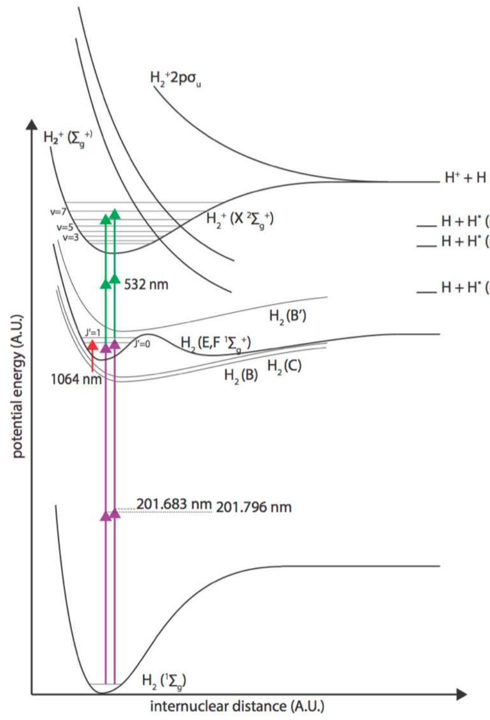
The observed behavior of H2 in this EF state demonstrate a very strong polarizability anisotropy. At higher energies investigated, the (E,F) states displays a alignment towards the main laser axis (cos² distribution), whereas at lower energies, it displays a tendency to follow a sin² distribution. This behavior is displayed even when energies are kept low for dissociation and only the alignment laser field is changed.

This behavior is attributed to emerge from Stark mixing J=2 rovibrational states to the prepared J=0 as the laser intensity increases. Quantification of these effects induces a polarizability anisotropy of H2 (E,F) of 302 ± 87 A.U. or 45 ± 13 Å³

Introduction

Similarly to atoms, molecules can also absorb more energy (such as photons) than required to ionize. Due to additional degrees of freedom, new process such as dissociative ionization and multiphoton ionization are observed.

Molecular hydrogen is the most abundant molecule in the universe, and despite being the simplest neutral diatomic, its behavior at higher energies remain a great theoretical challenge. Its exposition to intense laser fields can provide us ways to measure its properties. The (E,F) state of H_2 has been the subject of quite a number of studies in the past decade, as laser sources became more intense.

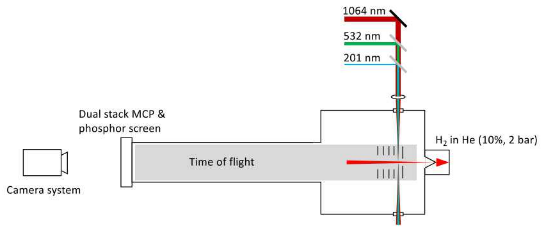


The (E,F) state of H2

The unusual shape of the (E,F) surface has two minima resulting from the avoided crossing of the E and F state potential energy curves. It is expected that the double-minimum nature of this intermediate state should manifest itself in several ways in the ionization due to the homogeneous perturbations between vibrational levels associated with the ionic inner (E state) and covalent outer (F state) wells and the expected large variation of the transition moment with an H–H internuclear distance.

Production of images

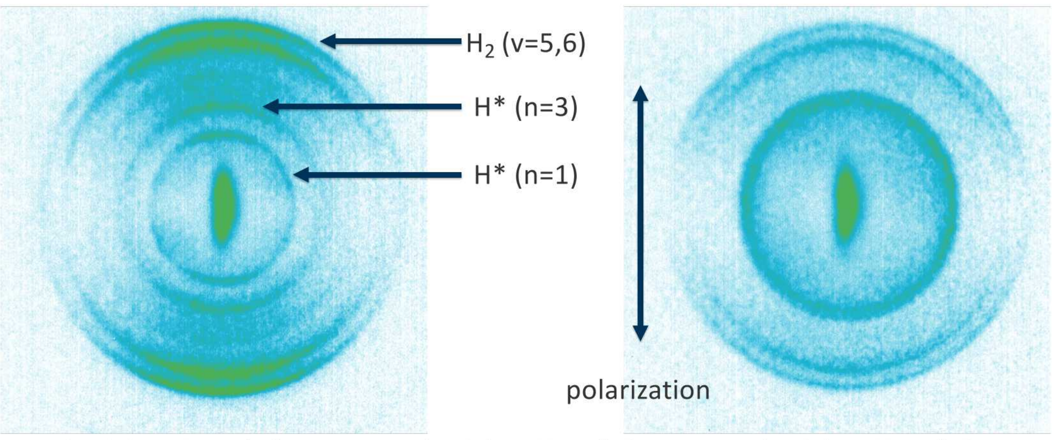
A molecular beam of 10% H_2 in He propagates from a pulsed molecular valve. A tunable ns laser at 201 nm generates the (E,F) state of H_2 in the desired J state. After 100 ns to allow for statistical reorientation of molecules, an intense 5-ns 1064 nm laser applies a field, whilst a 3-ns 532 nm laser dissociates excited H_2. The H_2^+ and H^+ are thereof extracted by the time of flight mass spectrometer. Ion impacts on the MCP induces a light on the position sensitive phosphor screen, recorded by the camera. A background image is then taken by imaging the background gas.



Analysis of the data

Several process are displayed and recorded on these images. Starting from the outer part:.

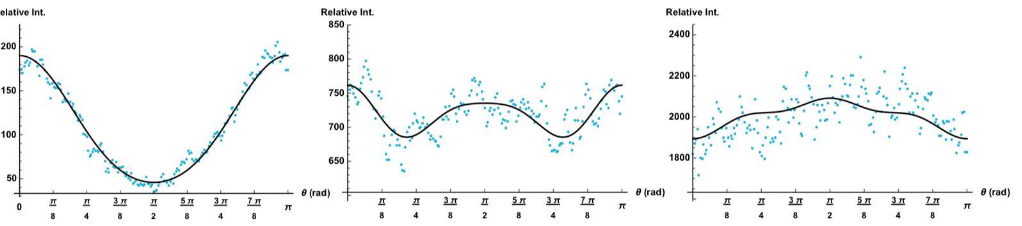
- H_2 ionization, from 2 x 532 photon absorption on the (E,F) state (v=5 and v=6 are visible on these images)
- H* (n=3) emerging from 2 different processes
  - H_2(E,F) + 2 x 532 nm > H (n=0) + H* (n=3)
  - H_2(E,F) + 4 x 1064 nm > H (n=0) + H* (n=3), statistically limited but we account for its presence
- H_2 (E,F) + 3 x 1064 nm > H (n=0) + H* (n=1), visible only on high energy (1064 nm) images.



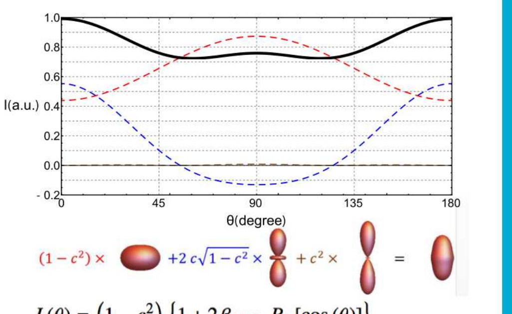
Dissociation: 5.27 MW/cm²
Alignment: 735 MW/cm2

Dissociation: 5.28 MW/cm²
Alignment: 160 MW/cm2

Dissociation: 5.28 MW/cm²
Alignment: 46.3 MW/cm²



From the images, we extract the intensity as a function of angle of the H* (n=3) population. This population peaks along the laser polarization axis for high laser intensity, but becomes round, and peaks orthogonally at lower intensities. Displayed here are the extreme cases (images and curve) and a curve for intermediate energy. The population distribution is now compared to expected distributions from pure molecular orbitals, to determine the mixing coefficient *c* for each image.



$$I(\theta) = \left(1-c^2\right)\left\{1+2\beta_{2(00)}\,P_2\left[\cos(\theta)\right]\right\} + c^2\left\{1+2\beta_{2(22)}\,P_2\left[\cos(\theta)\right]\right\}\left[d_{00}^2(\theta)\right]^2 + 2c\sqrt{(1-c^2)}\left\{1+2\beta_{2(02)}\,P_2\left[\cos(\theta)\right]\right\}d_{00}^2(\theta)$$

Displayed here are the pure functions and the mathematical representation of the intensity *I*.

We can express the population alignment $<\mathrm{Cos}^2(\Theta)> = {}^1\!/_3 + {}^2\!/_{135} \times (\alpha_\parallel - \alpha_\perp) \times \frac{E_0}{4B}$ at low field value, where α coefficients are the parallel and perpendicular polarizability components. The difference is Δα, polarizability anisotropy. E_0 is the electric field value, B the rotational constant of the (E,F) state.

With our data we obtain an anisotropy of Δα = 45 ± 13 Å³.
This value is the largest anisotropy measured to date, 4x as large as CS_2 (10 Å³)
A theoretical value of 956 Å³ exists for a static field, as opposed to our dynamic system. Are these two value compatible? More models are needed.