

INTRODUCTION

Insights into electron spin and nuclei is central to the field of chemical reaction dynamics. Of particular note is the study of spin-polarized hydrogen (SPH) atoms, which result from photodissociation of molecules. Examination of the detailed H-atom spin polarization is achieved by determining the projection of the electron spin onto the probe laser direction. In doing so, its angular distribution, complex dissociation pathways, and coherent excitation mechanisms may be revealed.

Approaches to detect SPH atoms are experimentally challenging due to the difficulty associated with probing ground-state H atoms through isolated fine structure levels, which is the only direct way to achieve sensitivity to the ground-state m-distribution.

Rakitzis et al have previously employed polarized fluorescence techniques in order to discriminate the fine structure-levels and subsequently detect SPH from the photodissociation of HCl and HBr. However, the unwieldy approach does **not** provide the angular dependence needed to determine distinct contributions to the polarization. In general, three parameters are necessary to describe electronic orientation. Two of these are produced by photolysis using circularly polarized light, and they generally vanish for dissociation of polyatomic molecules. The third parameter, γ_1' , is a coherent term induced by linear photolysis light and maximum at 45°.

Here, we present a novel methodology for **direct** detection of spin-polarized hydrogen atoms with high velocity resolution using a simple modification of the Hydrogen Rydberg time-of-flight technique. In doing so, a general probe of multi-surface nonadiabatic dynamics is achieved, sensitive to coherent effects in dissociation along multiple paths, and is applicable to a wide range of critical polyatomic systems.

PREVIOUS WORK

Contribution to the theory of the effect of orientation of atoms produced in photodissociation of molecules

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(Submitted 24 April 1981)
Zh. Eksp. Teor. Fiz. 80(10-1420) (1981)

A general theory is developed for the orientation effect of atoms produced in the photodissociation of molecules. The effect is studied qualitatively and quantitatively. Expressions based on the operators for the energy matrix of the excited state of a diatomic molecule are obtained and describe the population, orientation, and alignment of the atoms formed. The photodissociation of molecules, examples are given of molecules oriented by σ and π light. **POSSIBLE APPLICATIONS OF THE CONSIDERED PHENOMENON** required for the existence of the effect are discussed. **PHENOMENON** is shown to be observed in the case of the dissociation of molecules by linearly polarized light. It follows from the foregoing theoretical analysis that photodissociation of molecules by linearly polarized radiation makes it possible to obtain oriented atoms of such elements as **hydrogen, helium, neon, argon, krypton, and others**, many of which cannot be obtained by ordinary methods.

T.P. Rakitzis et al

- Fluorescence detection
- Detection is perpendicular to the laser axis and polarization is II

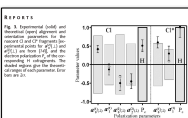


Spin-Polarized Hydrogen Atoms from Molecular Photodissociation

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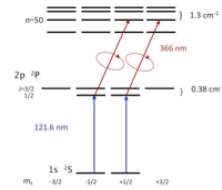
The production of spin-polarized hydrogen atoms from the photodissociation of hydrogen molecules with linearly polarized 200-nanometer light is observed from the measurement of the circular angular distribution of ground-state $2p_{1/2}$ and excited state $2p_{3/2}$ Ly-alpha emission. The experimental measurement of the circular angular distribution of the Ly-alpha emission and the circular angular distribution of the Ly-alpha emission are presented. The angular momentum distribution is a function of the photolysis laser polarization and the probe laser polarization.

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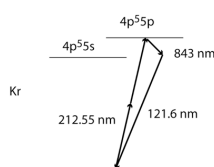
NEW APPROACH

Rydberg Tagging Spin-Selective Excitation Scheme



- Lyman-alpha excitation to $2P$ spin levels
- Possible to discriminate between $2P_{1/2}$ and $2P_{3/2}$ with narrowband polarized light
- Probe laser k-vector defines quantization axis
- Second photon from counter-propagating beam excites atom from $2P_{1/2}$ level to an $n > 50$ Rydberg state

VUV Generation

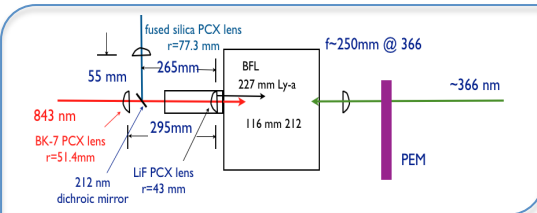


4-wave mixing

$$\omega_4 = \omega_1 + \omega_2 + \omega_3$$

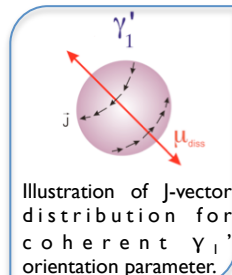
$$\omega_4 = \omega_1 + \omega_2 - \omega_4$$

$$\omega_4 = \omega_1 - \omega_2 - \omega_4$$

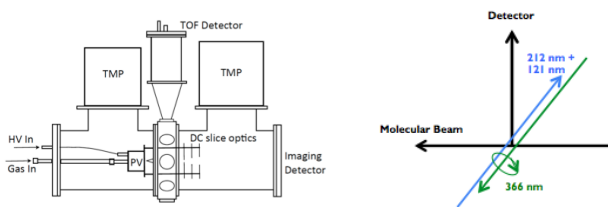
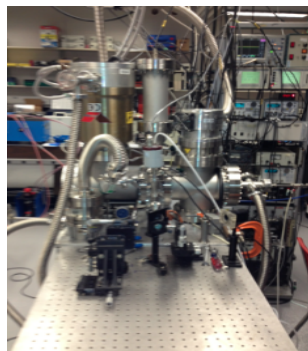


Theoretical Advantages

- Complete picture of photofragment orbital orientation described by **single anisotropy parameter, γ_1'**

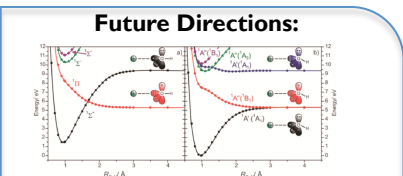


Experimental Scheme



- After ionizing, H-atoms fly to detector, ~100cm away
- TOF recorded by a high-bandwidth multichannel scalar card

Future Directions:



- H₂O, H₂S, and NH₃
- Acetylene
- E levels resolved in different resulting polarizations through rydberg tagging technique

Acknowledgements

