



## Editor's Choice

## Imaging detection of spin-polarized hydrogen atoms



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## ABSTRACT

We demonstrate that a two-color high- $n$  Rydberg excitation of hydrogen atoms using a circularly polarized tagging laser may be combined with pulsed-field ionization to allow for direct velocity map imaging of the H atom spin polarization. In this proof-of-principle study we only detect the incoherent spin polarization for a single velocity component in the 213 nm dissociation of HBr, but anticipate that by combining this strategy with slice imaging, the full velocity dependent H atom spin polarization may be obtained in a single experimental geometry.

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## 1. Introduction

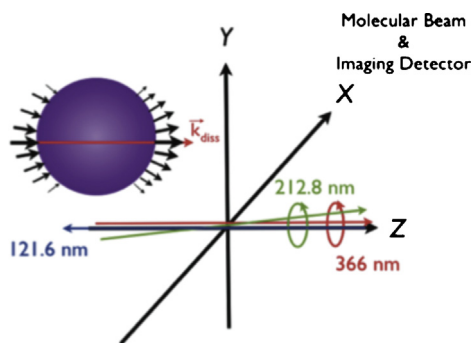
Photodissociation can give rise to hydrogen atoms with polarized electron spin. This phenomenon has long been predicted [1,2], and observed in other systems [3] but only in recent years have techniques to measure the H atom spin polarization emerged. The first such determinations were inferences based upon measurements of the cofragment angular momentum polarization for diatomic systems [4,5]. Subsequently, spin-polarized hydrogen (SPH) was directly measured under bulb conditions monitoring polarized VUV fluorescence, albeit with velocity resolution limited by a vacuum ultraviolet Doppler probe [6]. Very recently, we developed a direct probe of the velocity-dependent H atom spin polarization by adapting the H atom Rydberg time-of-flight technique [7,8]. This approach, relying on the two-color double resonance excitation of H atoms to high- $n$  Rydberg states, allows for spin-selective probe because a single fine structure component at the 2p level can be selected in the second 'tagging step', which then defines a laboratory reference for the spin projection. However, only two of the three parameters needed to fully characterize the H atom spin velocity distribution can easily be obtained in the conventional Rydberg tagging configuration [9,10] with the time-of-flight axis perpendicular to the laser propagation direction. In this report, we show that this same two-color excitation scheme may be adapted to an imaging configuration in which all three parameters can be obtained with their

velocity distributions. Again, the two-color double-resonance scheme allows us to isolate a single fine-structure component for spin-sensitive detection, but for the third incoherent parameter reported here, it will be necessary to scan the probe lasers and operate in a slice-imaging mode to record the full velocity distribution. In this letter we offer a proof-of-principle demonstration of the approach in which we measure this parameter at a single recoil velocity, as we have not yet implemented slice-imaging ion optics in the apparatus.

Probes of photofragment angular momentum polarization can give insight into details of the dissociation dynamics that may be inaccessible by other means [11–13]. This can include phenomena such as nonadiabatic transition probabilities, coherence transfer, and matter-wave interference. Hydrogen atoms are photoproducts for countless important molecules but to-date, only the hydrogen atom velocity distributions are usually measured. By simultaneously measuring the electron spin polarization along with the velocity distribution, we have the potential to increase the number of observables we may use to study these dissociation events. As a  $J=1/2$  system, H atom spin can exhibit orientation but not alignment. This spin orientation produced in photodissociation may be characterized by three anisotropy parameters,  $\alpha_1$ ,  $\gamma_1$  and  $\gamma'_1$  [14]. The first represents the incoherent contribution to the spin polarization produced by circularly polarized photolysis light, while the latter two are coherent contributions arising from circular or linear polarized photolysis light. In this report we demonstrate that the combined use of Rydberg tagging with velocity mapped imaging allows for direct determination of the  $\alpha_1$  parameter, permitting a complete characterization of the velocity-dependent H atom spin polarization. We note that these are lab frame anisotropy

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**Figure 1.** SPH-RTOF imaging experimental geometry.  $\alpha_1$  is obtained with circularly polarized photolysis, although all three parameters can be acquired in this arrangement; their unique angular distributions have been previously described [7,8]. In all spin-polarized H atom experiments, the Lyman- $\alpha$  beam is linearly polarized while the tagging probe is circularly polarized. Laser wavelengths are 121.6 nm for Lyman- $\alpha$ , 368.4 nm for the tagging laser, and 212.8 nm for photolysis. (For interpretation of the references to color in text, the reader is referred to the web version of this article.)

parameters, and they may readily be transformed to an alternative set of polarization parameters [15,16].

## 2. Experimental approach

A detailed description of the SPH-RTOF approach and apparatus has been reported in references [7,8]. Previously, we used the H atom Rydberg time-of-flight technique with linear or circular photolysis light to characterize two coherent reaction channels of H atom spin-polarization. We have now adapted the approach and the apparatus to do high- $n$  Rydberg *imaging*, which allows a more general probe of H atom spin polarization, albeit with somewhat reduced velocity resolution. The earlier time-of-flight configuration has been described in detail previously [7,8]; herein we include the experimental geometry for the imaging strategy, shown in Figure 1. In the earlier work we monitored the recoil distribution along the Y axis, perpendicular to the plane of the laser and molecular beams (referred to as Geometries I and II for circular and linear photolysis light, respectively). It is possible to obtain the coherent contributions in this case because for these two parameters, the spin polarization is maximal in this direction. However, as shown schematically in Figure 1, the  $\alpha_1$  contribution vanishes along the Y-axis but is maximal along Z, the photolysis (and probe) laser propagation directions. The imaging configuration detailed here allows for a direct measurement of  $\alpha_1$  as well as the coherent anisotropy parameters, by projecting the full spin-tagged recoil H atom distribution onto the detector as described in the following.

Our spin polarization measurements rely on a two-color double resonance excitation scheme described previously [7]. In this approach, photoproduct H atoms are excited to the 2p level using linearly polarized Lyman- $\alpha$  laser light. A second, counterpropagating ‘tagging’ laser that is right circularly polarized (RCP) excites the H atoms from the  $2p_{1/2}$  level to a high- $n$  Rydberg level, typically  $n=20$  or higher. Using the tagging laser propagation direction as the quantization axis, the selection rule for the first step (linearly polarized perpendicular to the quantization axis) is  $\Delta m = \pm 1$ , while for the RCP tagging step it is  $\Delta m = +1$ . Excitation is possible to the 20s or 20d levels, but the transition amplitudes to the former are small compared to the latter, and may be neglected. The result is essentially a pure spin tag of the photoproduct H atoms. In the time-of-flight case, these excited atoms are allowed to fly to a detector where they are field-ionized and counted. For the imaging configuration we simply wait 50 ns after the tagging laser and apply a pulse to the repeller and extractor of the velocity map imaging electrodes.

All of the tagged H atoms are ionized and velocity mapped onto the imaging detector 60 cm away. Softley and coworkers have successfully used a related hybrid Rydberg tagging/imaging strategy to effect slice imaging [17] and spatially-selective field ionization [18] of NO photoproducts, although they were not pursuing vector correlation or spin polarization measurements in that case. To obtain the full velocity distributions it is necessary to operate in a slice imaging mode [19,20], because in general we do not have the cylindrical symmetry necessary for inversion of the 2-D projections to the 3-D distributions. As mentioned above, however, our present apparatus is not currently equipped for high resolution slice imaging as needed for H atom detection. Nevertheless, by focusing exclusively on the limit of the velocity distribution scattered along Z, we can determine the value for the  $\alpha_1$  parameter for the single limiting recoil speed there. For this test case of HBr at 212.8 nm, this is in fact the dominant channel and the principal source of spin polarization [5,21]. The other point to emphasize is that to obtain the full velocity distribution for the incoherent contribution it is necessary to scan the frequencies of both the Lyman- $\alpha$  laser and the tagging laser together as discussed further below. However, for the present measurement at a single recoil velocity, laser frequency scanning is not necessary.

In this imaging approach, the pulsed field ionization occurs in the interaction region less than 50 ns after the tagging probe beam selects a single spin state. Considerable care is taken experimentally to ensure this brief ionization pulse occurs sufficiently long after the probe beam, as the presence of any significant fields in the interaction region will broaden the Rydberg transition, which would then no longer permit spin-selective detection. One can easily verify that broadening of a Rydberg transition has not occurred as a consequence of the pulsed-field ionization in the interaction region by scanning the tagging laser. Timing jitter of the ion optics pulsing ultimately determines how close the ionization pulse can be to the Rydberg excitation. If it is delayed too much then significant degradation of the velocity resolution is seen, as well as signal inhomogeneity arising from fly-out. It should be emphasized that all three parameters can be obtained in this ion imaging geometry; however, we here demonstrate measurement of the  $\alpha_1$  parameter that was inaccessible to our previous time-of-flight detection.

The H atom spin polarization is determined as follows (see Figure 1).

For Geometry I:

$$\frac{I_r - I_l}{I_r + I_l} = -\frac{3\sqrt{3}\gamma_1}{4 - \beta} \quad (1)$$

Geometry II:

$$\frac{I_r - I_l}{I_r + I_l} = -\frac{3\sqrt{3}\gamma'_1}{4 + \beta} \quad (2)$$

Geometry III:

$$\frac{I_r - I_l}{I_r + I_l} = -\frac{3\sqrt{3}\alpha_1}{2 - \beta} \quad (3)$$

$I_r$  and  $I_l$  refer to signal intensities obtained using RCP and LCP probe helicity, respectively. Geometries I and III employ RCP photolysis light, while Geometry II uses linearly polarized photolysis light at  $45^\circ$  from the Y axis. The expressions for Geometries I and II are given here for completeness, only results for Geometry III ( $\alpha_1$ ) are presented here.  $\beta$  is the conventional anisotropy parameter obtained for linearly polarized photolysis light. Finally, an important consideration in imaging spin-polarized H atoms is the fine structure selectivity. We must ensure our detection effectively discriminates between these transitions which are separated by only  $0.37 \text{ cm}^{-1}$ . One key aspect of the two-color scheme that aids in this is the differential Doppler selectivity for the two transitions. This helps to ensure that only a single 2p level is promoted to the high- $n$

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