

First Observation of Angular Momentum Orientation Transfer In Photodissociation of H₂

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In collisions of charged particles or photons with atoms, one generally probes the collision dynamics by measuring the number, energy, or angular distribution of the secondary or scattered particles. Additionally, one can study the polarization of the light emitted by excited post-collision fragments [1,2]. This provides detailed information about how angular momentum is distributed amongst the collision partners, allowing a deeper understanding of the collision dynamics. Light polarization studies can be particularly informative if the incident projectile, the target atom, or both are also polarized [3].



The perpetrators; from the left; Tim Gay, Kinte Allen, Joshua Machacek, John Furst, Kenny McLaughlin, Orhan Yenen

In the case of molecular targets, the angular momentum picture is complicated by two additional factors: the rotational angular momentum of the nuclei, and the fact that the orbital angular momentum of the electrons is no longer a good quantum number, due to the loss of spherical symmetry.

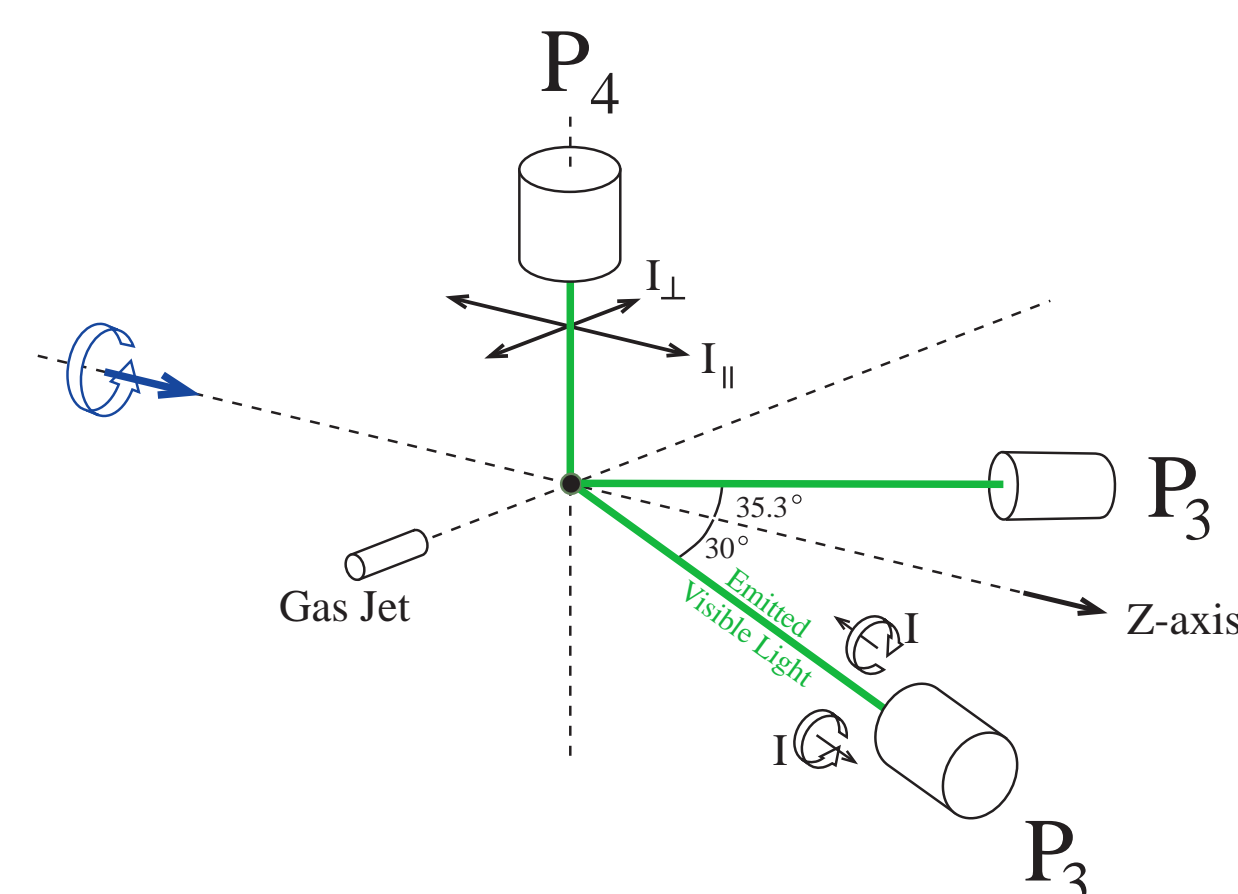
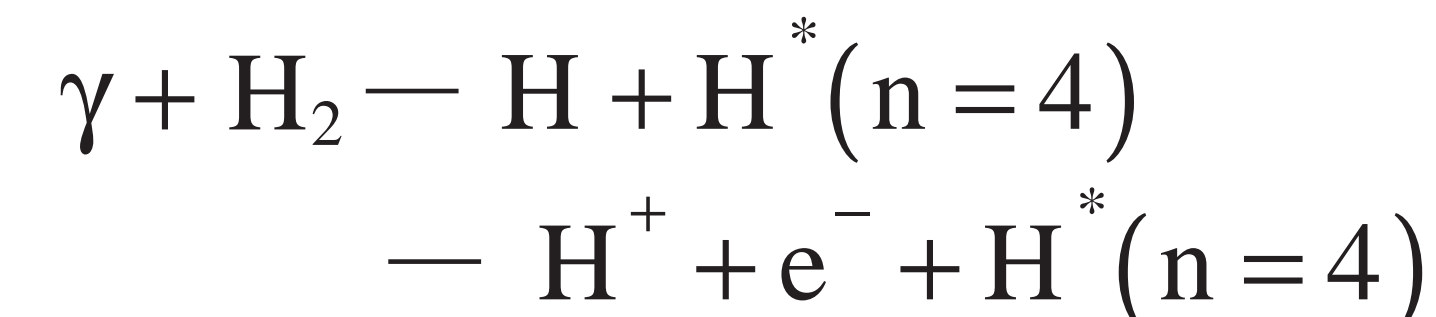


Figure 1: Experimental schematic.

Pioneering studies of photodissociation and photoionization of simple, diatomic molecules (see, e.g. references [4,5]) by photons showed the utility of measuring the fluorescence polarization. More recently, studies with linearly-polarized incident radiation have revealed unique interference phenomena when the angular momenta of the fragment atoms are determined using laser-induced fluorescence [6]. With the advent of third-generation synchrotron light sources such as the Advanced Light Source (ALS), high-resolution reactive

molecule-polarized photon collisions studies are becoming ever more feasible and, given the wealth of physical insight they offer into photoionization and photodissociation dynamics, increasingly important in molecular physics.

In an ongoing experiment at the Advanced Light Source, we are making the first studies of molecular photoexcitation and dissociation using circularly-polarized light in which the circular polarization of the photofragment fluorescence is studied. By measuring the circular polarization of the photofragments produced by circularly-polarized light, we obtain a direct measure of how efficiently angular momentum along the incident photon axis is transferred to the dissociated atoms. Initially, we have concentrated on the fundamental reactions in which the excited hydrogen atom subsequently emits H β (486 nm) radiation.



Linearly-polarized light from the 10.0.1.2 beamline passes through a four-reflection retarder that gives the light a degree of circular polarization > 99.7%. This light then enters a target chamber in which it crosses an effusive jet of H₂ gas with a number density of roughly 10¹⁴ cm⁻³ (see Figure 1). Light from the photofragments is monitored by three bi-alkali photomultiplier tubes at polar angles of 30°, 35.3°, and 90° relative to the incident beam axis. The 30° and 35.3° detectors measure the relative circular-polarization Stokes parameter P₃ [1]; the 90° detector measures the linear polarization fraction P₁.

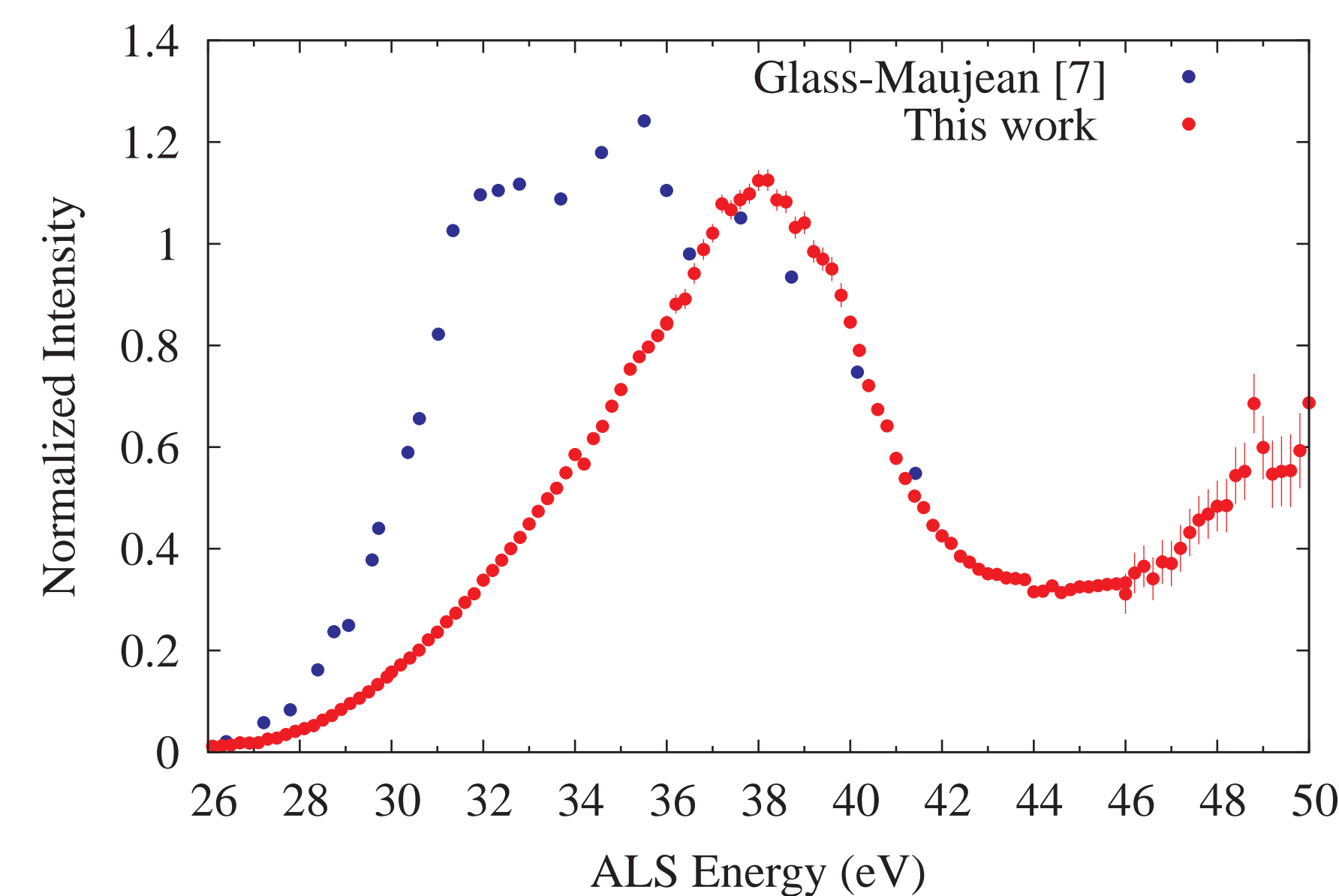


Figure 2: Measured fluorescence intensity.

Our preliminary results are shown in Figures 2 through 4, with photon energies ranging from 27 eV to 50 eV. The fluorescence intensity, which is proportional to the cross section for the reactions listed above, is shown in Figure 2. Our data is in significant disagreement with the only other available data: that of reference [7]. We have no explanation for this discrepancy. Our experiment is substantially similar to that of Glass-Maujean *et al.*, except that we used a narrow-band (10nm) interference filter to isolate the Balmer- β radiation, whereas they used a Wratten 40 filter [7]. The general shape of the 486 nm excitation curve can be understood by referring to a diagram of the so-called “super-excited” dissociative states of H₂^{*} (Figure 5 [8]). The maximum in the cross section corresponds to the excitation of the Q1 and Q2 bands [7]. Above 38 eV, however, these states are no longer accessible, and the threshold for Q3 and Q4 production is reached. We believe the rise in the cross section beyond 45 eV in Figure 2 represents the first direct evidence for the production of these higher-lying states.

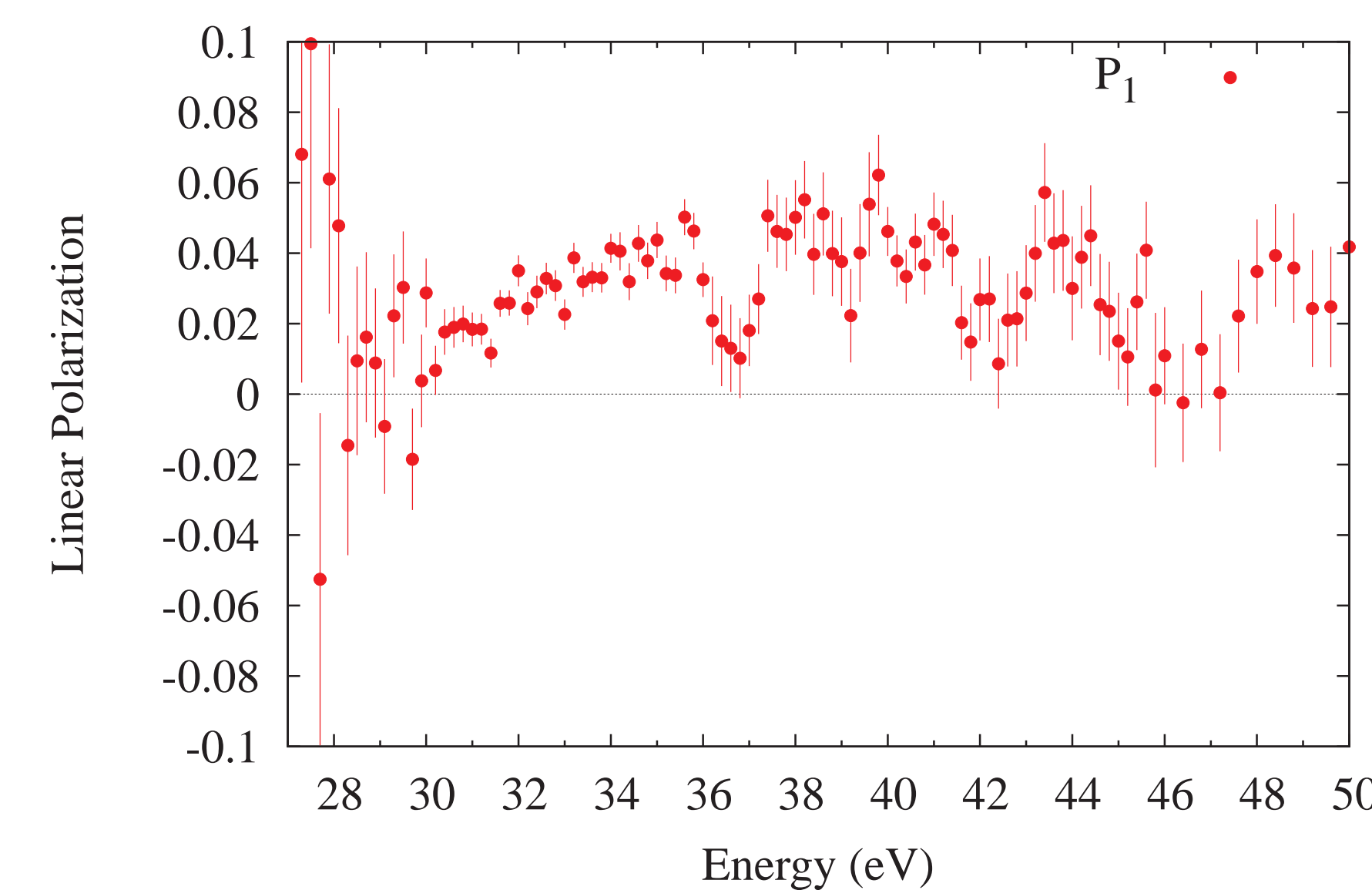


Figure 3: Linear polarization fraction P₁ vs. incident photon energy.

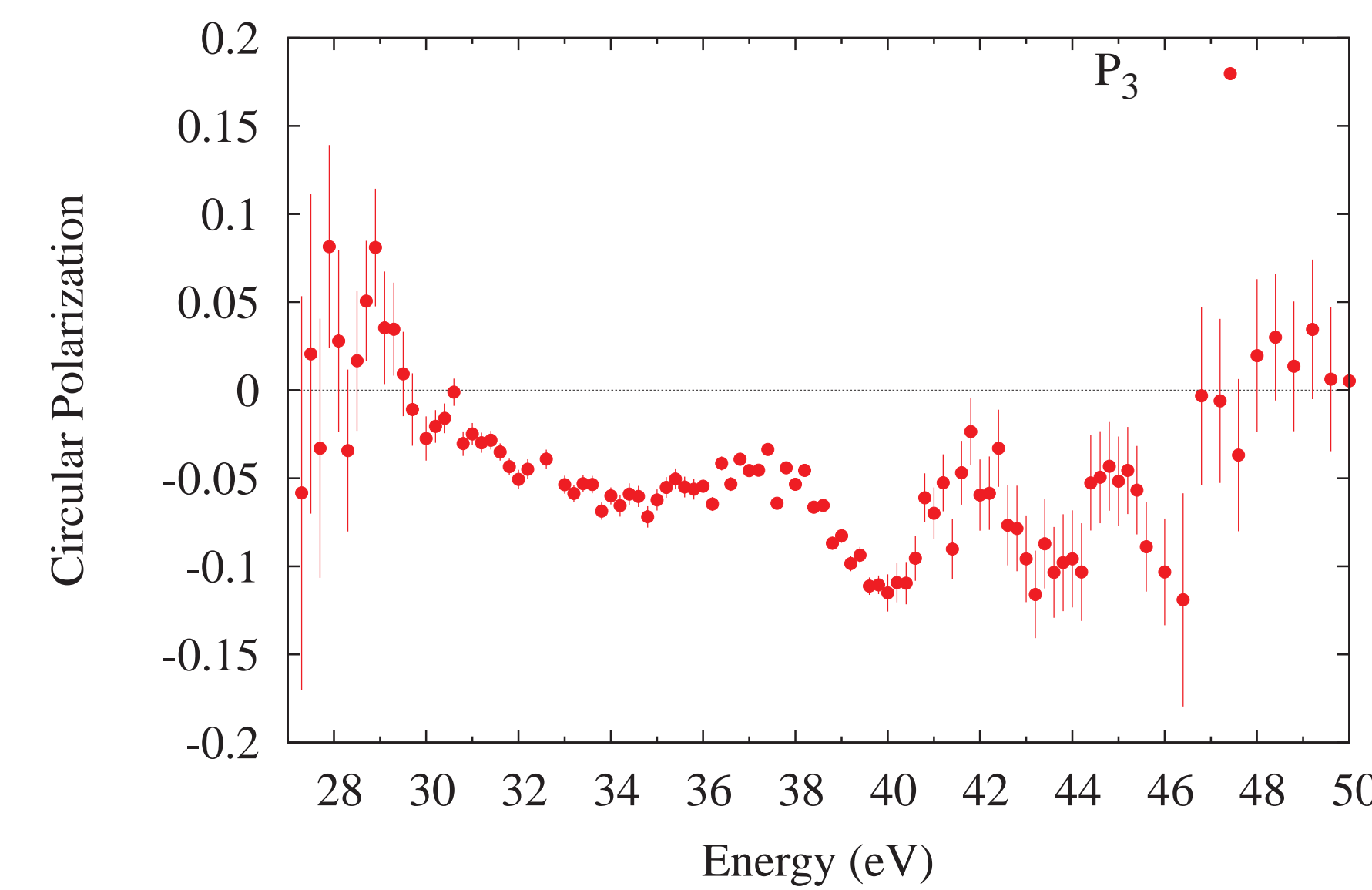


Figure 4: Circular polarization fraction P₃ vs. incident photon energy.

In contrast with the 486 nm production cross section, which is relatively smooth over energy scales of an eV or so, the polarization data exhibit significant structure. The reason for this structure is puzzling, given that the fluorescence we observe is the result of promptly dissociating states, as opposed to intermediate population of a predissociating state. The H β fluorescence we observe is the result of dissociation that occurs in less than 10⁻¹³ seconds, during which the internuclear axis is essentially fixed in space. Thus rotational or vibrational structure cannot explain the features we see. Even more intriguing is the hint of a sign reversal in the P₃ data at 29 eV. This corresponds to the radiating n=4 atoms having angular momenta that point oppositely to the angular momentum of the incident photons. Such a reversal is very surprising given the prompt destruction of the molecule.

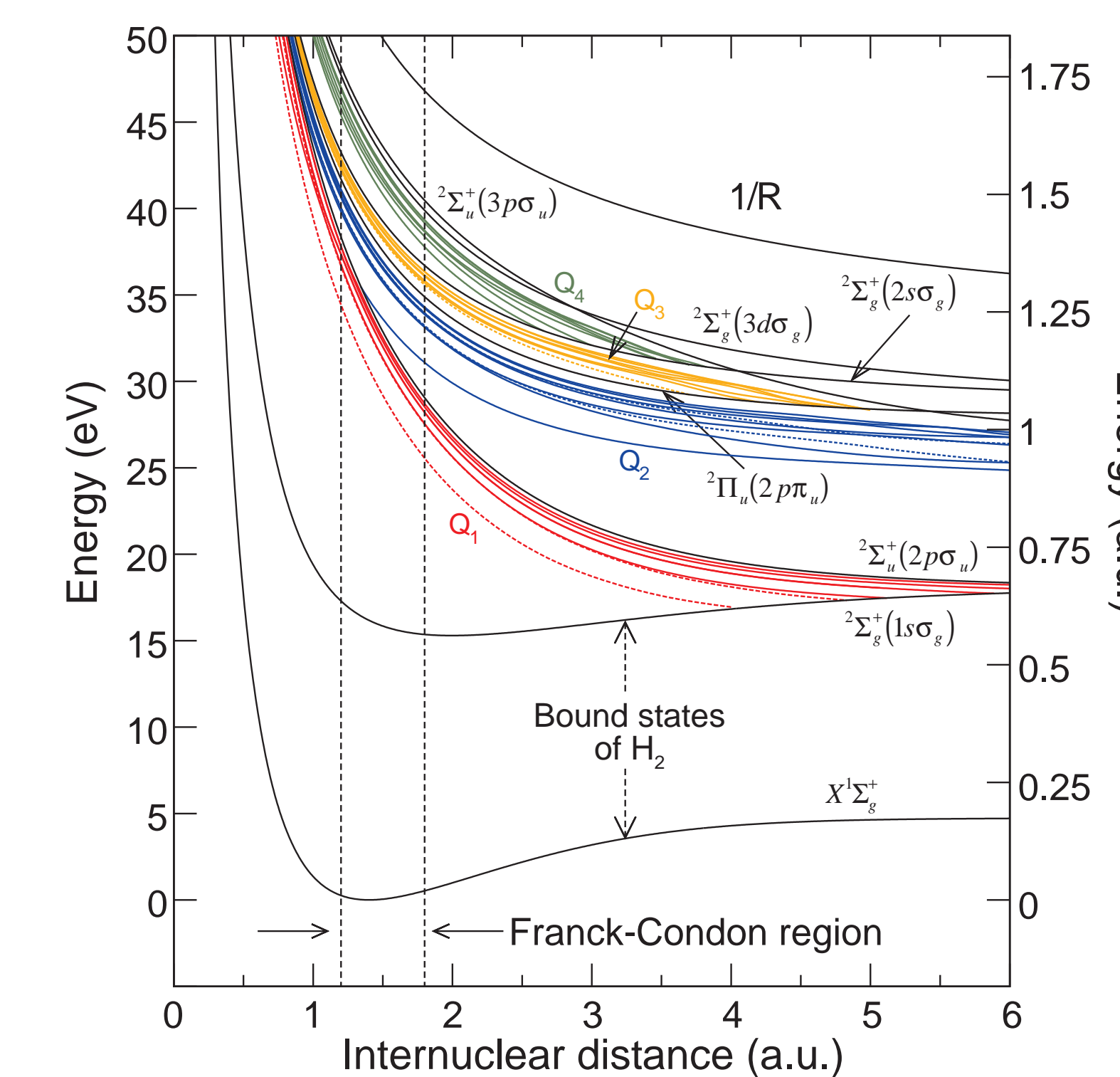


Figure 5: Potential energy curve of H₂ and H₂⁺ [8].

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- [1] U.Fano and J.H.Macek, *Rev.Mod.Phys.* **45**, 553 (1973).
- [2] K.Blum, *Density Matrix Theory and Applications*, 2nd ed. (Plenum, New York, 1996).
- [3] See, e.g., K.W.Mclaughlin *et al.*, *Phys.Rev.Lett.* **88**, 123003 (2002).
- [4] See, e.g., E.D.Poliakoff *et al.*, *Phys.Rev.Lett.* **46**, 907 (1981).
- [5] M. Glass-Maujean and J.A.Beswick, *Phys.Rev.A* **38**, 5660 (1988).
- [6] A.J.Alexander and R.N.Zare, *Acct.Chem.Res.* **33**, 199 (2000).
- [7] M.Glass-Maujean, *J.Chem.Phys.* **89**, 2839 (1988).
- [8] T.Aoto *et al.*, *Chem.Phys.Lett.*, **389**, 145 (2004).