

## Orientation of hydrogen $2p$ after tilted-foil excitation

C. J. Liu,\* T. J. Gay,<sup>†</sup> and K. P. Schüler

*Department of Physics, Yale University, New Haven, Connecticut 06511*

(Received 12 January 1989)

The circular polarization of Lyman- $\alpha$  photons resulting from beam-tilted-foil excited H( $2p$ ) was measured at energies between 9 and 20 keV with foil tilt angles up to  $60^\circ$ . Various interaction models are used to fit the data, and their physical significance is discussed.

### I. INTRODUCTION

It was pointed out independently by Fano and Macek<sup>1</sup> and Ellis<sup>2</sup> in 1973 that the breakdown of cylindrical symmetry resulting from tilting the foil could result in oriented magnetic sublevel populations of beam-foil excited atoms. Since then, elliptic polarization of optical radiation observed in both tilted-foil transmission<sup>3-10</sup> and grazing-incidence collisions<sup>11-19</sup> has received much attention, with the hope that it would lead to a better understanding of the atom-solid interaction. Experiments have established that the final surface interaction is dominantly important in beam-foil transmission collisions involving low- $Z$  projectile valence electrons. However, none of the various model calculations<sup>20-26</sup> that predict both orientation and alignment in beam-tilted-foil collisions agree completely with the experimental data accumulated so far.<sup>6,24-26</sup>

The excitation of H( $2p$ ) serves as a prototypical beam-foil collision for several reasons.

(1) The well-known wave functions of hydrogen significantly simplify *ab initio* calculations of the dynamical excitation process in the beam-foil interaction.<sup>24,26</sup>

(2) No complication due to the interaction of an oriented (or unoriented) core with the optically active electron<sup>25</sup> occurs.

(3) Since the proton cannot bind an electron inside a carbon foil at any velocity,<sup>27</sup> there will be no bulk effect to the observed excitation. This yields a direct test of the early surface electric field models<sup>21,23</sup> and Herman's anisotropic collision model.<sup>22</sup> Anisotropic electron pickup either from localized target states<sup>28</sup> or from the tail of the electron density at a metal surface<sup>24</sup> would be an important, if not dominant, orientation production mechanism for the tilted-foil-excited hydrogen atom.

(4) The postcollision Stark interaction after excitation is of a different nature in H<sup>0</sup> compared to He<sup>+</sup> or other hydrogenlike ions, since the long-range Coulomb surface electric field due to the ionic image charge is absent in the case of neutral hydrogen atoms. The short-range ( $\sim 5$  Å) dipole field of the bulk, however, is still important, especially because of the near degeneracy of H levels in a given  $n$  manifold. The Lamb-shift splitting for hydrogenic systems goes as  $Z^4$ . As a numerical example, the  $2^2S_{1/2}-2^2P_{1/2}$  Lamb shift in He<sup>+</sup> is  $\sim 14$  GHz, while it is  $\sim 1$  GHz for H<sup>0</sup>. It is, then, obvious that neutral hydrogen atoms are a unique species to study effects of

short-range surface electric fields due to the bulk in beam-foil interactions.

In brief, beam-foil-excited neutral hydrogen atoms provide the simplest system with which we can probe the complicated beam-foil interaction. The  $2p$  state is particularly interesting since it contains a relatively small number of substates but can still be oriented. Unfortunately, polarization analysis of Lyman- $\alpha$  (Ly- $\alpha$ ) emission is difficult because of its short wavelength.

The experimental arrangement and the polarization data for Ly- $\alpha$  emission of the H( $2p$ ) state resulting from beam-tilted-foil interaction are presented in Sec. II. Comparison of the circular polarization ( $S/I$ ) data with various models is discussed in Sec. III.

### II. EXPERIMENT

The data presented here<sup>29</sup> were obtained with the beam-foil polarimeter<sup>30-32</sup> that was developed at Yale to measure the proton polarization of the polarized H<sup>-</sup> source<sup>32,33</sup> at Brookhaven National Laboratory. The proton polarization of the ion source was turned off for the purpose of this work. After  $90^\circ$  bending by a static electric deflector, the H<sup>-</sup> beam was focused by an Einzel lens and magnetic quadrupole triplet, and steered by electric field plates into the target chamber where the pressure was kept below  $3 \times 10^{-6}$  torr.

Several carbon-foil targets of thickness  $5 \mu\text{g}/\text{cm}^2$  were mounted on inclined holders which accurately maintained foil-normal-tilt angles relative to the beam direction ( $\theta$ ) of  $0^\circ$  to  $60^\circ$ . A combined linear and rotary motion drive was used to translate the foil position with respect to the optical axis of a vacuum ultraviolet (vuv) polarimeter, thereby varying the effective time of flight of the atoms being observed. This drive was also used to swing each foil in and out of the beam path. The degree of circular polarization of the beam-foil generated Ly- $\alpha$  photons was analyzed by an optical polarimeter<sup>30-32</sup> consisting of a MgF<sub>2</sub> retardation waveplate,<sup>34,35</sup> a Brewster angle four-mirror linear polarizer,<sup>36</sup> and a Hamamatsu solar-blind photomultiplier with MgF<sub>2</sub> window and CsI-overcoated photocathode.<sup>32</sup> The analyzing power of the optical polarimeter was calibrated and known to be unity within 4%.<sup>32</sup> Optical components of the vuv polarimeter were preceded by a  $3 \times 5\text{-mm}^2$  rectangular slit that sampled a well-defined segment of the beam. The MgF<sub>2</sub> re-

tardation plate cuts off wavelengths shorter than 1150 Å and the quantum efficiency of the CsI photocathode drops off rapidly at wavelengths longer than 1700 Å. Thus the optical train conveniently isolates 1216 Å Ly- $\alpha$  light. Another photomultiplier, identical to the one in the optics chamber but without polarimeter optics in front of it, is used in the target chamber for beam current normalization. The source was operated in a pulsed mode at a repetition rate of  $\sim 1$  Hz when the data were taken. Beam intensity at the foil was nominally 10  $\mu$ A

with a beam pulse width of  $\sim 450$   $\mu$ s. The counting rate was  $\sim 16$  counts/pulse at a foil-tilt angle  $60^\circ$ . Typically, a data point with a statistical accuracy better than 1% was obtained in ten minutes.

In the data analysis, the effect of hyperfine interactions on the 2p manifold following excitation was taken into account. The fine-structure beats ( $\sim 10$  GHz) are averaged out by the wide collimating slit (3 mm) and need not be considered. The time dependence of the observed circular polarization is thus given by<sup>32</sup>

$$\frac{S}{I}(t) = \frac{\langle L_x \rangle}{\hbar} \frac{43 + 8 \cos(\omega_{1/2}t) + 5 \cos(\omega_{3/2}t)}{48 + [\frac{3}{2}(\rho_{1,-1} + \rho_{1,-1}^*) - \frac{1}{2}(\rho_{1,1} + \rho_{-1,-1} - 2\rho_{0,0})][5 + 3 \cos(\omega_{3/2}t)]}, \quad (1)$$

where  $\langle L_x \rangle = \sqrt{2}\hbar \text{Re}(\rho_{1,0} + \rho_{0,-1})$  is the initial angular momentum of the electron cloud along the viewing axis, the  $\rho_{ij}$ 's are the components of the  $3 \times 3$  2p orbital angular momentum density matrix,  $\omega_{1/2}$  and  $\omega_{3/2}$  are the hyperfine splittings of the  $j = \frac{1}{2}$  and  $j = \frac{3}{2}$  2p levels, respectively. In the case of complete orientation along the viewing axis, i.e., if  $\langle L_x \rangle = \hbar$ ,  $S/I$  is 100% at  $t = 0$  as expected. The calculated  $S/I$ , assuming negligible cascading downstream from the foil, an ideal collimating slit of zero width, and complete orbital orientation at  $t = 0$ , is illustrated in Fig. 1. Taking the spatial resolution of the apparatus due to finite collimator width and cascading from the 3D (assumed to have negligible initial orbital orientation) and 3S states into account, the time dependence of  $S/I$  with the foil tilted at  $60^\circ$  is shown in Fig. 2. Initial populations of the 3D and 3S states were taken to be 15% and 35.5% of the initial 2p population, respectively.<sup>32,37</sup> The zero of the time scale was taken to be such that the downstream edge of the foil was centered in the slit. In the experiment and in these calculations, the

long axis of the slit is perpendicular to the beam axis.

The circular polarization ( $S/I$ ) of the Ly- $\alpha$  photons as a result of beam-tilted-foil interaction at 20-keV beam energy is shown in Table I. The value of  $S/I$  quoted herein has been obtained by multiplying our observed values of  $S/I$  by a correction factor, which was taken as the ratio of  $S/I$  in Fig. 1 and the calculated  $S/I$  as in Fig. 2 for various tilt angles. The downstream edge of the foil was assumed to be centered in the slit with an accuracy better than 1 mm. The contribution of photons reflected from foil surface into the detection system is believed to be negligible. As a test,  $S/I$  were measured with the foil translated 2.38 mm upstream and 1.62 mm downstream from its nominal position. The observed  $S/I$  agreed well within statistical accuracy with the calculated time dependence of  $S/I$ . The applied correction factors were  $1.08 \pm 0.03$ ,  $1.07 \pm 0.03$ ,  $1.06 \pm 0.02$ , and  $1.04 \pm 0.04$  for foil-tilt angles at  $\pm 60^\circ$ ,  $\pm 45^\circ$ ,  $\pm 30^\circ$ , and  $\pm 15^\circ$ , respectively. As seen in the Table I, the circular polarization reversed sign, as expected, when the foil was tilted in the

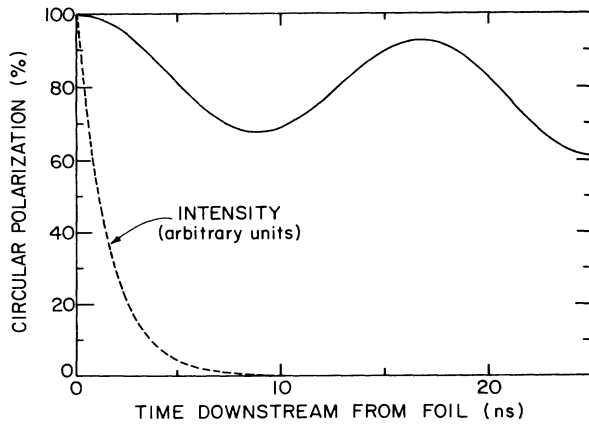


FIG. 1. The calculated hyperfine quantum beats in  $S/I$  assuming complete initial orbital orientation  $\langle L_x \rangle = \hbar$ , negligible cascading from higher excited states, and ideal collimating function [see Eq. (2)]. The rapid falloff of the Ly- $\alpha$  intensity is also shown in this figure as a reference.

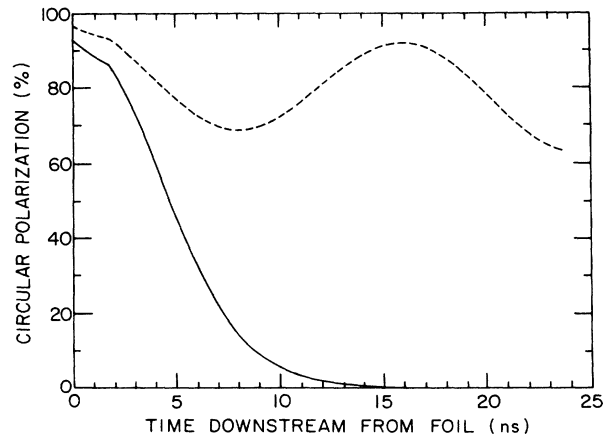


FIG. 2. The solid line shows the expected time dependence of  $S/I$  with both the slit function and cascading from 3D and 3S taken into account. The short dashed line shows the hyperfine beats with slit function taken into account only.

TABLE I. Measured circular polarization vs foil-tilt angles  $\theta$  at 20-keV beam energy. Positive angles are for the cases in which the foil normal was tilted counterclockwise relative to the incident beam direction, while negative angles are for clockwise rotation.

$\theta$ (deg)	$S/I$ (%)	$\theta$ (deg)	$S/I$ (%)
60	$15.5 \pm 0.7$	-60	$-15.2 \pm 0.7$
45	$11.1 \pm 0.6$	-45	$-11.8 \pm 0.5$
30	$7.9 \pm 0.7$	-30	$-7.3 \pm 0.5$
15	$4.1 \pm 0.4$	-15	$-3.4 \pm 0.6$

opposite sense.

The energy dependence of orientation was also observed for foil-tilt angles of  $30^\circ$  and  $-60^\circ$ , with incident beam energies between 9 and 20 keV. These data are shown in Table II (see also Fig. 3). The expected energy loss in the foil at each energy and tilt angle was calculated using proton stopping power in carbon<sup>38</sup> at the corresponding energy and effective thickness of the foil at each tilt angle.

### III. DISCUSSION

Various models were tried to fit the experimental data (see Fig. 4). The results are shown in Table III.

It is noted that the apparently linear relation of the observed circular polarization versus tilt angle can be resolved into two components,  $\sin\theta$  and  $\sin 2\theta$ , which correspond, respectively, to the first- and second-order Stark effect<sup>21,23</sup> of the surface electric field on the emerging atoms. The fitted function (at 20 keV)

$$S/I(\theta) = 21.0 \sin\theta - 3.3 \sin 2\theta \quad (\text{in } \%), \quad (2)$$

indicates that the orientation within the context of the surface electric field model comes mainly from torque on the nonzero electric dipole moment<sup>39</sup> rather than from transformation of the alignment,<sup>20</sup> which is very small over the measured energy range.<sup>40</sup> It is also noted that

TABLE II. Energy dependence of the circular polarization ( $S/I$ ) at foil-tilt angles  $30^\circ$  and  $-60^\circ$ .

Tilt angle (deg)	Incident beam energy (keV)	Circular polarization (%)	Expected <sup>a</sup> energy loss (keV)
30	19.3	$8.5 \pm 0.8$	$\sim 2.89$
30	18.0	$7.9 \pm 0.5$	$\sim 2.78$
30	17.0	$7.9 \pm 0.5$	$\sim 2.71$
30	15.0	$6.0 \pm 0.5$	$\sim 2.57$
30	13.0	$7.0 \pm 0.5$	$\sim 2.41$
30	11.0	$5.1 \pm 0.5$	$\sim 2.22$
30	9.0	$5.8 \pm 0.5$	$\sim 1.99$
-60	19.3	$-15.2 \pm 0.7$	$\sim 5.00$
-60	11.0	$-8.1 \pm 0.5$	$\sim 3.85$

<sup>a</sup>Reference 38.

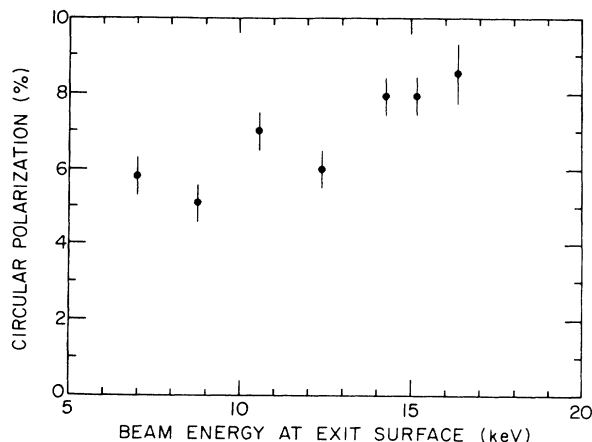


FIG. 3. Energy dependence of the circular polarization with foil normal tilted at an angle of  $30^\circ$  with respect to the beam axis.

various model calculations within the context of the density gradient model<sup>24,28,41</sup> also predict a  $\sin\theta$  dependence of  $S/I$ . A  $\sin\theta$  dependence has recently also been observed by Winter and Ortjohann<sup>42</sup> who used an  $H^+$  beam of 70 keV and a wide range of foil-tilt angles. They obtained a good fit with  $S/I = 22 \sin\theta$ , measured in %, which describes foil data taken in transmission and in reflection geometries. Cascading corrections were not given, but were likely to be small in their case. It appears that the somewhat higher circular polarization observed by Winter and Ortjohann at 70 keV follows the trend of the energy dependence seen in our data (see Fig. 3). Data

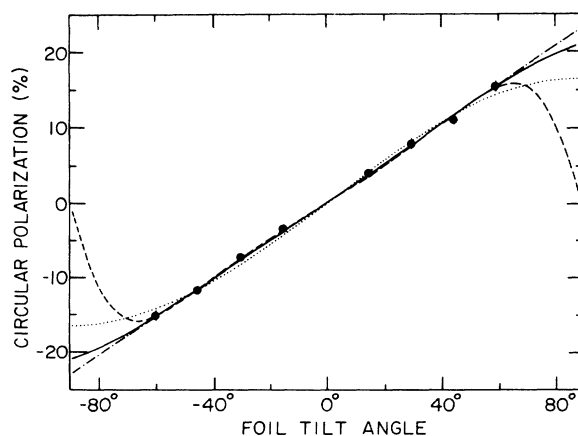


FIG. 4. Circular polarization ( $S/I$ ) of  $Ly-\alpha$  radiation resulting from beam-tilted-foil interaction. The circles represent the experimental data. The dotted, dashed-dotted, short dashed, and solid lines in the figure represent the best fit to the density gradient (and/or torque) model, the linear model, Band's, and Lombardi's surface electric field model calculations, respectively, as explained in Table III.

TABLE III. Parametric fits to the circular polarization data. The last column gives expressions for  $S/I$  (measured in %) with  $\theta$  (measured in deg).

Model	No. of data points	No. of parameters	$\chi^2_v$	$S/I$
Linear	8	1	0.4	$0.26\theta$
Torque <sup>a</sup>	8	1	1.5	$16.5 \sin\theta$
Lombardi's <sup>b</sup>	8	2	0.4	$21.0 \sin\theta - 3.3 \sin 2\theta$
Band's <sup>c</sup>	8	3	0.6	$14.5 \tan\theta$
				$1 + 0.12/\cos^2\theta - 0.08 \cos 2\theta/\cos^2\theta$

<sup>a</sup>Both the torque model (see Ref. 5) and the density gradient model (see Refs. 24, 28, and 41) predict a  $\sin\theta$  dependence.

<sup>b</sup>Different model calculations of the surface electric field model (Ref. 21).

<sup>c</sup>Different model calculations of the surface electric field model (Ref. 23).

on the energy dependence of the electric dipole moment along the beam axis are available.<sup>43</sup> Extensive measurements on the energy dependence of the  $\sin\theta$  coefficient would provide information about the validity of the surface electric field model versus the density gradient model.

It is important to mention Winter and Ortjohann's data on the polarization of Ly- $\alpha$  photons after the interaction of H<sup>+</sup> ions with the monocrystalline Ni surface at a grazing angle of about 0.8° in UHV conditions ( $\sim 10^{-11}$  mbar) (Ref. 19). The reported  $S/I$  of the Ly- $\alpha$  emission was very high ( $\sim 50\%$ ) with beam energies be-

tween 15 and 29 keV. Extrapolating the foil data presented here to high tilt angles based on the various fitting functions does not yield a value of 50% for  $S/I$  at 89.2° tilt angle. Also, the already mentioned recent foil data of Winter and Ortjohann at 70 keV (Ref. 42) do not approach such large values. The observation seems to indicate there is a fundamental difference between the beam-tilted-foil transmission interaction and ion-beam-surface scattering at grazing incidence.<sup>13</sup> Another possibility is the material or texture dependence of the final surface interaction, which can be studied under ultrahigh vacuum conditions ( $\sim 10^{-10}$  torr).

\*Present address: Physics Division, Argonne National Laboratory, Argonne, IL 60439.

†Present address: Department of Physics, University of Missouri, Rolla, MO 65401.

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