ALIGNMENT OF HIGH RYDBERG STATES IN HYDROGEN

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Abstract

We have measured the light yields and polarizations of the light emitted from several Balmer transitions in atomic hydrogen following beam foil excitation of protons at energies of 50 to 150 keV. The polarizations have been measured as a function of distance downbeam from the exciter foil for several transitions. The measurements indicate a very strong initial alignment which is then perturbed by surface fields out to several mm from the surface.

INTRODUCTION

Using a standard beam-foil arrangement, we have observed the fluorescence from the Balmer series in atomic hydrogen for principal quantum numbers $n=4$ to $n=15$. The initial intent of the measurements was to study the shapes of the wavefunctions of high Rydberg states after foil excitation and to check some of the earlier suggestions made from observations of electric-field stripped electrons from such states (1,2). Simple electron capture theory predicts populations of states proportional to n^{-3} . (From a simple density of states argument: $E \propto -n^{-2}$, leading to $dE/dn \propto 2n^{-3}$). However, some observations at lower n , and for other beam-foil excited systems, indicate different population dependences on n , which are sometimes not monotonic (3,4). Observations of the polarization of the emission indicate asymmetries in the shape of the excited state wavefunction. Typically, for lower n states, beam-foil excitation leads to wavefunctions which tend to be elongated along the beam axis - that is, the $m=0$ state is more populated than the $m \neq 0$ states (the beam axis being the z -axis of quantization). In figure 1 we show states where only $m=0$ is populated, for successively higher angular momentum values ℓ . Clearly, in general, such a population distribution corresponds to a cigar-shaped distribution along the beam axis. The opposite case, of a pancake-like wavefunction, with axis of rotational symmetry along the beam axis, corresponding to high m populations, is only rarely seen in low n -state excitation; the only examples being for hydrogen $n=2$ (5), and for some d-states in He I (6).

In this work, we have measured the linear polarization for the $n=4$ up to the $n=7$ Balmer lines as a function of the distance from the exciter foil. Once the atom reaches the field-free region away from the foil surface, the polarization shows quantum beats induced by the spin-orbit fine structure. These well-understood beats can be used to indicate where this field-free region begins. For example, since the

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Fig. 1 Wave functions showing complete alignment along the beam-axis, with only $m=0$ populated.

spin-orbit interaction is internal to the atom, the total light yield is unaffected. This is in contrast to an external field which can change both the polarization observed, and the light yield in both a single direction and that into all directions. We have used this feature to draw a very surprising conclusion from our data: the higher n states, $n=5$ to 7 , are strongly affected by external fields out to distances of up to 10 mm from the foil surface. Clearly, higher n states, for example $n=20$ to 50 , which are observed in the field-stripped electron measurements, will be even more strongly perturbed by these external fields. We suggest an origin for this surface electric field. However, such a field does not explain all the observations.

EXPERIMENT

Spectra were taken over the short wavelength range shown in Fig. 2 to obtain the relative yields for high n -states $n=9$ to 14 . Within this $100\text{\AA}$, the relative photon collection efficiency remains constant to within $\pm 10\%$. The grating efficiency decreased to higher wavelengths, while the quantum efficiency of the photomultiplier increased, both by about 4% within this range.

Fig. 2 Spectrum of high n Balmer transitions from 100 keV foil excited hydrogen.

The polarization measurements were made using a standard polarimeter (7). Since only perpendicular foils were used in these measurements, only the two linear polarization components, $I(\text{par})$ and $I(\text{perp})$, were measured. An example of these results is shown in Fig. 3. Four measurements of each polarization component were made at each foil position. The foil was stepped up-beam to record the complex decay curve.

Measurements were made using H^+ , H_2^+ and H_3^+ projectiles at energies between 50 and 120 keV. The molecular projectiles were dissociated by a pre-foil, generally located about 6 mm in front of the exciter foil. The entire target chamber was surrounded with a mumetal shield to reduce the earth's magnetic field. Without shielding, the decay curves from the higher n -states were strongly perturbed.

RESULTS

First we fit the total light yields for the different n states. For the integrated profiles of $n=9$ to 14, we obtained a power law dependence on the principal quantum number of $-(7.01 \pm 0.3)$. In this range of n , the lifetimes except for n_s states, scale approximately as $n^{-3.1}$. Thus, the populations scale as $n^{-3.9 \pm 0.3}$. This is somewhat faster than the scaling law predicted from the density of states model. The next step is to analyze the quantum beat curves to find the beginning of the field-free region for each of the decays.

Fig. 3 Decay curves of $n=2-5$ observed in light polarized parallel and perpendicular to the beam axis. Note the changes in x-scale near the foil. Each set of 8 channels are separated by 10 times the distance beyond channel 92 (from 0.10 to 1.0 mm).

In Fig. 4 we show the two decays for polarizations parallel and perpendicular to the beam for the $n=2-6$ Balmer lines. After about 1 cm from the foil the beats are clearly out of phase by 180 degrees, with the parallel polarization beats of double the amplitude of the perpendicular polarization beats. This is indicative of a field-free region, where the total light yield, $I(\text{tot}) = I(\text{par}) + 2 \cdot I(\text{perp})$, shows no quantum beats, and should be a simple multi-exponential decay. We show this in Fig. 5, where the $n=4, 5, 6$ and 7 total light yield decays show clearly the beginning of the field-free region. The higher n states show stronger anomalous intensities near the foil since their ℓ sub-states are more strongly mixed by the surface electric field. A qualitative measure of this mixing is given by the ratio of intensities between the first maximum and the first minimum of the total yield decay curves. This function increases strongly with n from about 1.1 for $n=4$ to 5.5 for $n=7$.

Fig. 4 Decay curves for the $n=2-6$ Balmer transition observed in polarizations parallel and perpendicular to the beam axis. The x-scale change occurs at channel 128.

The onset of the field-free region occurs at distances from 7 to 15 mm from the foil surface. This is much larger than the few hundred angstroms expected. The field-affected distance varies only slightly with ion-beam current or with foil thickness between 2 and $20 \mu\text{g}\cdot\text{cm}^{-2}$.

We have made Fourier transforms of the polarization quantum beats in the field-free regions for the Balmer transitions from $n=4, 5, 6$, and 7. An example of the $n=4$ curve (Balmer beta) is shown in Fig. 6. Four frequencies are expected as indicated on the energy level diagram of Fig. 7. These are found as indicated. They are observed with similar relative intensities for the higher n states, except that the lowest frequency beat of the $d_{3/2} - d_{5/2}$ fine structure is not measured for the $n=6$ and $n=7$ curves (its wavelength being longer than the observation region). Including the parts of the polarization curves within the field-affected region, of course, washes out these observed frequencies in the Fourier transforms.

Figure 5. Total light yields, $I(\text{par}) + 2 \cdot I(\text{perp})$, for the $n=2-4$, 5, 6, and 7 transitions, as functions of distance from the foil. Note the change in x-scale near the foil. The foil surface is at the first open circle. Field-free decay begins at the second open circle.

Fig. 6 Fourier transform of the $n=4$ polarization observed in the field-free region. The peaks A to D correspond to the fine structure splittings labelled in Fig. 7.

Fig. 7 Energy levels of an n -state in hydrogen indicating the fine structure beats seen in the field-free balmer transitions.

CONCLUSIONS

We find that the light yields from high Rydberg states in beam-foil excited hydrogen indicate strong surface electric field mixing out to about 1 cm beyond the foil surface. The mixing is stronger for the higher n states. This surface field reduces a very strong initial alignment at the surface, where the population appears to be almost completely in the $m=0$ states (z -axis along the beam-axis). At the beginning of the field-free region, the alignment is still positive - a high $m=0$ state population, but strong mixing of the s and d states is observed. The depth of the surface field is also shown by the unusual phase of the $P_{1/2} - P_{3/2}$ quantum beat, which is not 0 or 180 degrees relative to the foil surface, as would be expected for a sharp turn-off of the surface field at the foil surface.

Singer et al. (8) have previously proposed an extended surface electric field to explain previous observations of quantum beats in beam-foil excited Balmer beta. We suggest that these measurements confirm the presence of this field and may be used to map out its extent and other properties. Due to the finite conductivity of the thin carbon foil, which we measure to have a diametrical resistance of a few megohms, an electron charge is built up on the foil surface by the ion beam. The electric field is due to this electron surface distribution surrounded by the circular foil holder at ground potential about 3 mm from the beam center. This macroscopic electric field will thus be of the order of a few hundred volts per cm at the center dropping to zero at the foil edge, and also to zero at some equivalent distance along the beam. Our results do not conform to all the characteristics of this field. Most significantly, when we decrease the foil resistance (by increasing the foil thickness), or increase the electron production (by increasing the ion beam current), only small changes in the decay curve within the field-affected region occur. We are developing a model to describe the time development of the mixed states travelling through the extended surface field.

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