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Observation of coherent excitation processes in e-H collisions*

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In a measurement of emission of Balmer-α flux from atomic hydrogen or deuterium excited by electron impact in a crossed-beam experiment, a longitudinal electric field which enhances the Balmer-α flux by Stark-mixing components of like M_J within the $n = 3$ shell, does so asymmetrically depending on whether the field is applied parallel or antiparallel to the electron beam. The effect is attributed to coherent excitation of states of opposite parity.

INTRODUCTION

Measurements of cross sections for electron-impact excitation of the $3S$, $3P$, and $3D$ states of atomic hydrogen, from observations of Balmer-α radiation, were recently reported by this laboratory.[1] These measurements were sensitive to ambient electric field strength through Stark mixing, and a careful study of this sensitivity was carried out. In the course of that investigation we observed a strong asymmetry in the electric field dependence of the Balmer-α flux[2,3] for electric field parallel or antiparallel to the electron beam axis. This asymmetry is predominately due to electric field mixing of coherently excited $3P$ and $3D$ states. A detailed theory for this electric field dependence has since been given by Mahan[3] and by Krotkov.[4] Here we report the relevant features of the measurements; discuss the basic physical ideas; and report the measured field dependences, comparing them with those calculated[2,3] from Born-approximation excitation amplitudes. This comparison is made for data obtained at 500-eV impact energy, the highest energy at which the asymmetry was measured and an energy regime where the Born approximation largely satisfies the usual validity criteria.

EXPERIMENTAL CONFIGURATION

The measurements were carried out in a high-vacuum crossed-beam apparatus previously described.[1,5] Figure 1 is a schematic representation of the interaction region in the plane determined by the horizontal electron-beam axis and the vertical axis of observation. An atomic hydrogen (or deuterium) beam passed perpendicularly through the intersection of the other two axes.

The electron gun was capable of electron-beam currents of approximately 50 μA at the 500-eV electron-beam energy used in the present measurements. The half-width of the electron-beam profile in the interaction region was typically 0.75 mm as determined by observations of transition radiation emitted from a solid-silver beam probe.[6] The electron-beam energy resolution was approximately 0.3 eV.

The chopped, collimated atomic beam had an essentially square umbra and penumbra, 6.7 and 9.1 mm on a side, respectively. The atom density at this plane was about 6×10^8 cm^{-3}, in a 90% dissociated beam. The molecular component contributes about 2% to the total Balmer-α signal,[3] which is neglected for the present measurement. Background gas pressure was ~6×10^{-8} torr.

The principal components of the detection optics were two $f = 50$ mm, $F/1.2$ collimating lenses; a 1.5-nm-FWHM interference filter centered at 656.3 nm, the Balmer-α wavelength; a cooled GaAs photomultiplier; and a rectangular light pipe which provided light averaging over the photocathode. Pulse amplification and counting techniques were used.

The electric field imposed on the interaction region was obtained by applying a voltage V_G to the annular electrode labeled G in Fig. 1 which guards the Faraday cup. An analog computation of the potential distribution confirmed that the field was approximately uniform and parallel to the electron-beam axis within the volume of overlap between the electron and atom beams. The potential gradient of up to ±20 V/cm imparted an energy component to the electrons in the interaction region which was compensated by adjusting the cathode potential of the electron gun.

EXPERIMENTAL RESULTS

Figure 2 shows results obtained at 500-eV electron energy. The Balmer-α intensity on the optical axis is shown for electric field strengths up to 20 V/cm applied parallel and antiparallel to the electron-beam axis. The error bars on the data points represent one standard deviation, plus an uncertainty in the 2–3% correction for the effects of space-charge fields, obtained by extrapolating

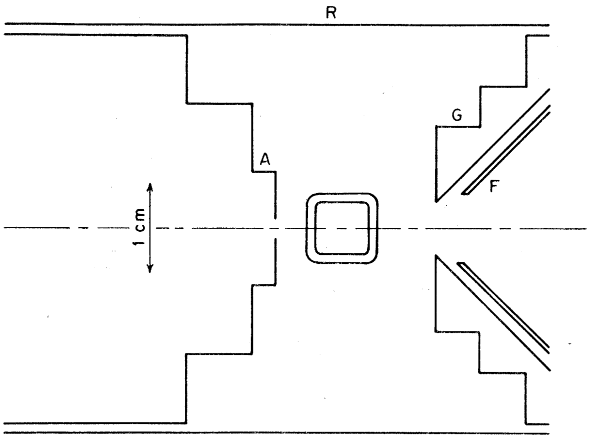

FIG. 1. Schematic representation of the cross section of the interaction region in a plane containing the electron beam axis and perpendicular to the atom-beam axis. The electron beam is horizontal and directed to the right, while the Balmer-α radiation is detected through a screen-covered window at the top of the diagram with a detector that is insensitive to polarization. The various electrodes have axial symmetry on the electron-beam axis. Electrodes A and R are electrically grounded. F, the Faraday cup, is normally grounded. In the present work electrode G is electrically biased in order to apply electric fields in the region of intersection of the electron-beam axis with the atom beam, for which the approximately square umbra and penumbra are outlined.

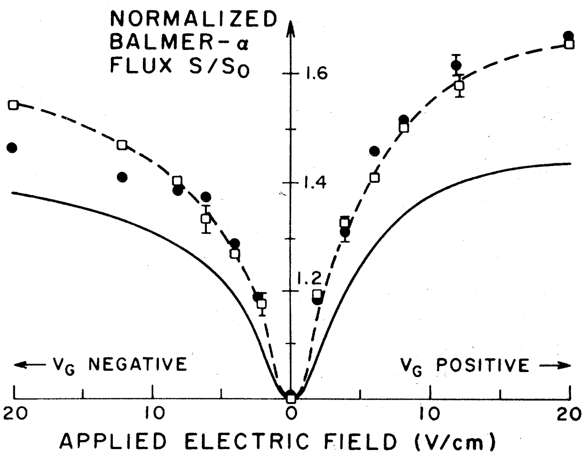

FIG. 2. Measured ratio of Balmer-α emission at the indicated field (S) to that at zero field (S_0) from (□) atomic hydrogen and (●) atomic deuterium excited by impact of 500-eV electrons, as a function of axial electric field. The dashed curve is a representation of the atomic-hydrogen result. The solid curve is from the Ref. 3 calculation, which uses two-state mixing equations and Born cross sections, neglecting hyperfine structure and cascading. The results of Ref. 4, which uses fewer approximations, are essentially identical. The ordinate gives the total signal in units of the zero-field signal. The measurements plotted to the right side of the ordinate axis were obtained with a positive voltage on electrode G (see Fig. 1); those to the left, with a negative voltage.

the data points individually to zero electron current.

The Balmer-α intensity rises rapidly from the zero-field value as the applied electric field increases in either direction, and it saturates at sufficiently high applied fields. Of interest is the asymmetry in the Balmer-α response to fields of the same magnitude applied in opposite directions. An accelerating field (conventionally a negative field, taking the direction of the electron velocity as positive) is seen to produce a larger Balmer-α response than does the retarding field of the same magnitude. At an electron-beam energy of 500 eV, an applied field of magnitude 20 V/cm produced an asymmetry in Balmer-α flux from deuterium which is about 17% of the zero-field signal. A corresponding asymmetry obtained at an electron-beam energy of 200 eV was about 21%. Results obtained using atomic hydrogen (with larger hfs) were about 11% and 17% at 500 and 200 eV respectively.

Possible instrumental asymmetries were carefully examined. The influence of applied fields upon the electron-beam and upon the photon counting rate was studied. The total electron current to the grounded Faraday cup was independent of the applied field to within 1%. A large positive voltage applied to the Faraday cup did not increase current collection. This confirmed complete collection of the electron beam and the absence of secondary electron currents. The integrating property of the light pipe minimized spatial photocathode efficiency correlations.

Because of the slope of the Balmer-α excitation cross section as a function of energy, the maximum field applied, if uncompensated, would have reduced the Balmer-α asymmetry by only about 4% of the zero-field flux. This component of the asymmetry was recovered by compensation with the cathode voltage. Subsequent analysis showed that the compensation used was about 50% too large, leading to an overstatement of the maximum observed asymmetry by slightly less than 2%. This 2% overstatement is not corrected in Fig. 2, but the values cited in the text do include a correction.

Finally, the extrapolation to zero electron current was found to contribute equally to the Balmer-α signal with the applied field parallel and antiparallel.

INTERPRETATION

An electric field applied along the electron-beam axis mixes opposite parity sublevels having the same magnetic quantum number M_J. (Hyperfine coupling is discussed below.) Since the $3P$ levels are more heavily populated in the process of excitation by 500-eV electrons than are the $3S$ and $3D$ levels, the net effect of this mixing is to increase the $3S$ and $3D$ populations at the expense of the $3P$ level population. Because only one-ninth of the radiation arising from decay of the $3P$ level is seen as Balmer-α radiation, the predominant fraction going into Lyman-β radiation, this shift in population increases the total Balmer-α intensity.

A conventional treatment of Stark mixing for atoms initially in a single state (e.g., following excitation) yields population changes that are symmetric in the electric field E and proportional to E^2 for small fields. An incoherent superposition of initial populations thus also yields a symmetrical dependence on the sign of an applied electric field. If one generalizes to allow for excitation into an initial superposition of states having some preferred initial relative phases, then the intensity calculations may include interference terms which have a linear dependence on the field strength, which is necessary for the occurrence of a directional asymmetry.

To test the validity of this interpretation one of us (A.H.M.) has carried out calculations[3] of the Balmer-α emission resulting from mixing of the three pairs of $n=3$ states L_{J,m_j} of opposite parity and with the smallest energy separations ($P_{3/2,3/2}-D_{3/2,3/2}$, $P_{3/2,1/2}-D_{3/2,1/2}$, and $S_{1/2,1/2}-P_{1/2,1/2}$) since these dominate the asymmetry and mixing at the experimental fields. Figure 3 shows the energy-level structure of the $n=3$ shell.

The formulation of the problem followed that of Percival and Seaton,[8] but in Mahan's work their two-step process of excitation and spontaneous emission was modified to include Stark mixing of pairs of states. Born excitation amplitudes were used, and the quantum-mechanical phases were carefully preserved through each step of the excitation, mixing, and spontaneous emission processes. The anisotropic nature of the radiation from the mixed fine-structure levels is also included in the formulation. Krotkov independently calculated[4] the mixing of all $n=3$ states with the same M_J. The results of both calculations are very similar for the experimental fields. Each predicts an asymmetry which is in the direction of that observed experimentally, but the theoretical asymmetry is smaller than the observed, in each case amounting to about 6% of the zero-field intensity. Cascade contributions from higher states were omitted from both calculations, but these should contribute little to the asymmetry, since they provide less than 10% of the total $n=3$ populations.

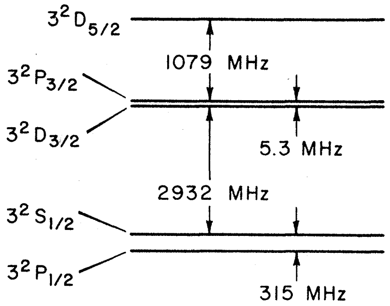

FIG. 3. Energy intervals within the $n=3$ shell of atom hydrogen (Ref. 7).

Hyperfine effects have also been omitted, which is only appropriate if the hfs splittings are less than the natural radiative widths.[8] This is more nearly satisified for deuterium, so that hfs should have less influence in that case. The inclusion of hyperfine structure will further dilute the initial LM_L coherent excitation, and diminish the predicted E dependence and asymmetry, which is consistent with the difference observed in hydrogen and deuterium. A more detailed calculation, enlarged to include hyperfine structure, could certainly be carried out. However, one could estimate the major effect of hyperfine structure on the asymmetry observed in I_{90}, the fluorescence perpendicular to the electron beam, by noting that the inclusion of new levels into the decay primarily reduces the polarization of the Balmer-α radiation. A calculation of the E dependence for totally depolarized fluorescence was carried out by Mahan; the calculated asymmetry in I_{90} at 20 V/cm dropped from 6% to 4.5% of the zero-field intensity. The expected asymmetry should lie somewhere between these limits. The discrepancy between experimental results and theoretical prediction is thus slightly larger than indicated by the comparison of 6% predicted to 17% measured for deuterium. This presumably derives from an inadequacy of the Born approximation to predict the coherences at 500 eV in spite of its expected accuracy for total cross sections. The observed coherence results from an integration, over scattered electron direction, of the phase relations between different $n=3$ state amplitudes. Thus it probes a quite different aspect of the excitation than that tested by total cross sections.

In summary, the observed asymmetry is seen as a manifestation of coherent excitation of super-

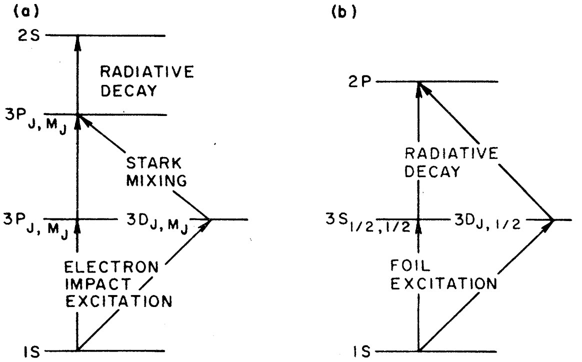

FIG. 4. (a) Illustrates the basis for the asymmetry observed in the present work from coherent excitation of two states of opposite parity followed by Stark mixing of one into the other. (b) Illustrates the basis for zero-field quantum beats as observed in beam-foil spectroscopy (Ref. 9), from radiative decay of coherently excited states of the same parity.

position states of the $n=3$ shell in atomic hydrogen (or deuterium). Interference terms may arise whenever any two substates can decay to the same lower state. In the present case, the significant interference terms arise from coherent excitation of states of *opposite parity*, one mixed into the other by the applied field and only that one carrying a Stark-mixing coefficient with its linear dependence on electric field [Fig. 4(a)]. This is in contrast to the interference effects which are observed as zero-field quantum beats in beam-foil spectroscopy.[9] These occur, in the absence of a mixing field, from coherent excitation of two excited states of the *same parity*, and angular momenta differing by two units, which are then allowed to decay to the same lower state [Fig. 4(b)].

Invoking experimental axial symmetry, a simple consideration of the electronic charge density distribution of superpositions of states of the same and of opposite parity[10,11] is informative. The former represents a quadrupole-like distortion of the atom which retains symmetry with respect to the beam direction. The latter does not retain this symmetry, but represents a net displacement (dipole) of the electronic charge cloud with respect to the nucleus. For the latter case, this picture provides plausibility for the idea that electric fields in opposite directions along the experimental axis should affect the electronic charge density distribution differently.

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