Observation of Coherent Electron-Density-Distribution Oscillations in Collision-Averaged Foil Excitation of the $n = 2$ Hydrogen Levels*

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Using foil-excited H beams (80–400 keV) we have applied a simple technique recently proposed by Eck to unambiguously separate genuine excitation coherence from that induced by fine structure and probe fields. We find compelling evidence for strong excitation coherence and have studied quantities related to the collision-averaged s-p phase coherence angle. Using Eck's theory, the data exhibit coherent fore-aft oscillations of the electron cloud with respect to the proton.

In a recent Letter,[1] Eck proposed a particularly simple technique for unambiguously separating collision-averaged foil-excitation coherence of H atoms from that induced by fine-structure interactions, and by the probe fields required to couple levels of opposite parity, which otherwise do not decay to the same final state. Basically, the technique depends on the use of electric probe fields $\vec{E}$ respectively parallel and antiparallel to the beam to exploit the fact that the excitation coherence signal is odd under field reflection, whereas the other signals are not.

A number of investigations[1-3] have dealt with observable excited-state coherence in simple systems (H, He, Li^+) induced by the fine-structure interaction, when initial magnetic-substate population asymmetry (alignment) prevails. The observations do not demonstrate true excitation coherence, since axial symmetry and the Russell-Sanders–coupling approximation require that only states of the same m_L and m_S can be coherently excited, while states of different J but the same L can still interfere because of the fine-structure interaction. The latter oscillations have been clearly described by Macek[2] and by various other authors, most recently Alguard and Drake.[3] Bashkin and Beauchemin[4] appear to be the first to have observed such oscillations with foil targets.

Apparently, only one other experiment[5] lays claim to observation of coherence between states of different L, in which possible s-d beats in the foil excitation of H_β were noted. These interesting experiments differ from those reported here in that levels of the same parity were involved, permitting one to consider quadrupole rather than dipole oscillations, but requiring the fitting of six unknowns to the total H_β intensity to extract various oscillatory components of light emitted from the large number of upper levels concerned. That fitting ambiguities result is demonstrated by the fact that the best fit to the H_β data gives appreciable initial phase angles[5] for the $p_{1/2}$-$p_{3/2}$ and $d_{3/2}$-$d_{5/2}$ fine-structure beats, in contrast to Macek's prediction of pure cosinusoidal oscillations.[2] A very slight foil-location uncertainty could easily have produced this result.

In the present experiment, only two unknowns (the amplitude and phase of difference signals uncer electric field reversal) require fitting, and the number of upper levels involved in Lyman-α emission is so small that interesting inferences can be drawn regarding initial state populations and their coherence properties. Moreover, the opposite parity of the Lyman s and p levels permits interpretation in terms of dipole distortions of the initial electron charge cloud, as well as the development of these distortions in time.[6]

A simple physical picture of the relationship between true excitation coherence and field reversal is as follows. If there is an initial displacement of electron charge cloud with respect to the proton, or one develops in time because of an inequality in proton and average electron axial velocity, the displacement will be either enhanced or diminished depending on the direction of $\vec{E}$ relative to that of the charge displacement. Incoherent coupling and quenching effects, light-intensity anisotropies, etc., depend on $|\vec{E}|$, but not on whether $\vec{E}$ is parallel or antiparallel to the quantization axis defined by the common axis ($+z$) of the beam and $\vec{E}$. For $\vec{E}$ perpendicular to the beam Eck's arguments[1] show that coherently excited s and p states are not coupled.

Eck's expression[1] for the beat signal is

$$\left(\frac{V}{\omega}\right)^2\left[\tfrac{1}{3}(\sigma_{p0}+2\sigma_{p1})-\sigma_s\right]\cos\omega t+\frac{1}{\sqrt{3}}\frac{V\omega_0}{\omega^2}\langle|f_s||f_{p0}|\cos\alpha\rangle\cos\omega t+\frac{1}{\sqrt{3}}\frac{V}{\omega}\langle|f_s||f_{p0}|\sin\alpha\rangle\sin\omega t, \qquad (1)$$

plus terms smaller by a factor $\Gamma/2\omega$, all exponentially damped with a damping constant $\Gamma/2$. Here, Γ is the average of perturbed s- and p-state decay rates, ω_0 is the Lamb shift, ω is the perturbed $s_{1/2}$-$p_{1/2}$ level splitting ($\equiv\omega_0$ for $|\vec{E}|=0$), V is the dipole matrix element $(s_{1/2}|-e\vec{E}\cdot\vec{r}|p_{1/2}) = -\sqrt{3}\,eEa_0/\hbar$, $t=0$ at the foil, and the excitation amplitudes for s- and p-state excitation are $f_s = |f_s|\exp(i\alpha_s)$ and $f_{p0}=|f_{p0}|\exp(i\alpha_p)$, with $\sigma=\langle|f|^2\rangle$ and $\alpha\equiv\alpha_s-\alpha_p$. The angular brackets refer to collision averages. We use this expression and hence the two-state and other approximations used by Eck, except that a better value for ω is obtained by diagonalizing the 3×3 perturbation matrix, incorporating the $p_{3/2}$ state ($\sim 10\omega_0$ away in energy). Hence the sum of the signals yields the incoherent oscillations superposed on nonoscillating perturbed $s_{1/2}$, $p_{1/2}$, and $p_{3/2}$ decays, while the difference yields only the coherent excitation terms, containing the s-p phase coherence angle α.

Figure 1 displays the sum signal (top curve), difference signal (bottom curve), and also a field-free decay curve through which a line representing a theoretical field-free p-state Lyman-α decay curve has been drawn. All curves are uniformly normalized, and actual photon counts are shown. Both sum and difference oscillations of comparable magnitude clearly occur. Apart from downstream cascade effects just starting to become appreciable at large t, the field-free decay curve matches theoretical expectations, neglecting the small, geometry-attenuated, zero-field intensity oscillations studied by Dobberstein, Andrä, and Wittman.[7]

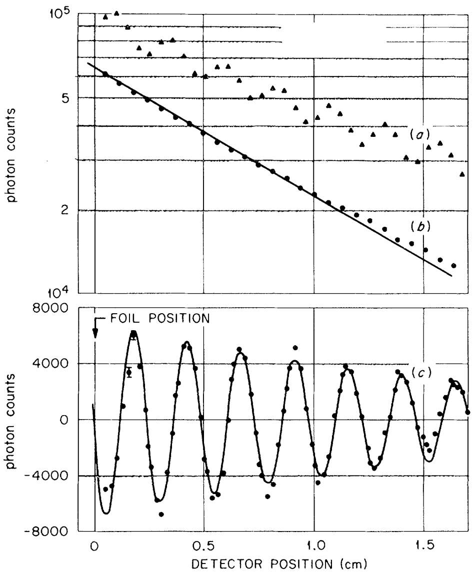

FIG. 1. Variation of signal strengths with distance downstream at 525 and 0 V/cm and 186 keV beam energy. The raw data for zero field are shown with an arbitrarily normalized straight line superposed, whose slope corresponds to τ_{2p} (b). The sum of the signals for $\vec{E}$ parallel and antiparallel to the field is plotted in the top, curve a, and the difference signal ($E_{\text{par}} - E_{\text{antipar}}$) is plotted in the bottom, curve c.

The experiment was performed by passing a 0.080-in.-diam, 2-deg-divergence, 186-keV proton beam (after the energy-loss correction) through a ~ 15-μg/cm^2 foil forming the upstream boundary of a parallel-plate condenser oriented perpendicular to the beam, whose symmetry axis coincided with that of the plates within ordinary machining tolerances. Fields of alternate polarity were established by measuring voltages applied to the downstream plate at 0.7 in. in relative separation with a calibrated differential voltmeter. Signals were obtained using a BX762 Bendix encapsulated Channeltron in perpendicular viewing geometry, sensitive between 1150 and 1900 Å and with peak sensitivity near Lyman α. The signal detector viewed the radiation through a movable pair of vertical 15-mil straight-edged slits, 0.25 in. tall, located about 2.75 and 5.1 in. from the horizontal beam. The finite trapezoidal field of view caused wave-form averaging, producing an effective ratio of observed wave-form amplitude to actual of about $\frac{3}{4}$. Normalization of signal strength per incident particle was accomplished by a second BX762 monitor counter viewing a section of beam along the third orthogonal direction through a pair of $\frac{1}{4}\times\frac{1}{4}$-in.^{2} slits. Phase shifts due to exponential damping of the wave form coupled with finite-length viewing geometry amounted to ~ 1 deg and were negligible for present purposes. Foil location was measured to within 5 mil. We used a variety of field strengths (66–1050 V/cm) and beam energies (80–400 keV). Sum and difference oscillations were always observed, though of course the larger oscillations obtained for the higher field strengths yield better data.

The solid curve drawn through the difference oscillations shown in Fig. 1 results from fitting only the coefficients of the $V\cos\omega t$ and $V\sin\omega t$ terms in Eq. (1). All other quantities ($V = 7.31$ GHz, $\omega = 15.27$ GHz, $\Gamma = 0.31$ GHz) were calculated from first principles and fundamental data. The resemblance to a suitably damped, negative-going sine wave of the correct frequency is evident. Because $\omega_0/\omega \sim 0.44$, the relative size of the $\cos\omega t$ difference terms is suppressed by this factor. Hence our lower-field data (still under analysis) will yield better information about limits on the size of $\langle |f_s||f_{p0}|\cos\alpha\rangle$ than the data of Fig. 1. What is clear from Fig. 1 is that $\langle |f_s| \times |f_{p0}|\sin\alpha\rangle$ has a *positive* value, since V is intrinsically negative, and the wave form matches Eck's prediction very well. The sum oscillations exhibit a minimum near the foil, in agreement with the conclusions of Alguard and Drake,[3] who used perpendicular fields at somewhat different energies.

It is not obvious that one can factor $|f_s||f_{p0}|$ out of $\langle |f_s||f_{p0}|\sin\alpha\rangle$, since $|f_s|$, $|f_p|$, and α can all vary in a correlated way from collision to collision. It is somewhat remarkable, then, that the collision averaging permits survival of the coherent terms. If one makes the simplifying assumptions that $|f_s|$ and $|f_{p0}|$ can be factored out of both terms, and in addition, α has a unique average value $\overline{\alpha}$, then the data of Fig. 1 suggest that $\sin\overline{\alpha} > 0$, and $\cos\overline{\alpha} < \sin\overline{\alpha}$. If $\overline{\alpha}$ were 0 (or π) for example, the coherent part of the initial wave function would involve $|f_s|u_s \pm |f_{p0}|u_p$, corresponding to a concentration of the electron charge distribution in the backward (or forward) hemisphere, resulting from the $\cos\theta$ dependence of u_p. Here u_s and u_p are the field-free spatial eigenfunctions of the $2s$ and $2p$ states. If $\overline{\alpha}$ were near $\pi/2$, however, or $\sin\overline{\alpha} \sim 1$, then the initial wave function would involve $i|f_s|u_s + |f_p|u_p$, corresponding to zero *initial* charge-distribution asymmetry, but *one which would reach peak concentration in the backward hemisphere at the subsequent time* $t = \pi/2\omega$, *and in the forward hemisphere at* $t = 3\pi/2\omega$. Thereafter, the charge-distribution asymmetry would continue to ring periodically between the two limits. For $\overline{\alpha}$ intermediate, some combination of initial distortion and initial "velocity" asymmetry would prevail. The data of Fig. 1 suggest that $\overline{\alpha} \sim \pi/2$; a better value awaits the low-field data analysis and improvements on the assumptions already noted.

Eck[6] has pointed out the complete analogy between these charge-distribution asymmetries at $t = 0$ and thereafter, and the conditions on the initial position and velocity of a classical oscillator. In effect, the physical content of the difference oscillations in Fig. 1 is the suggestion that while the initial dipole charge distribution may be fairly small, the initial collision-averaged electron cloud "velocity" is *not* small, and furthermore the initial cloud "velocity" *lags* that of the proton, since using Eck's theory, the electron concentration initially grows in the backward hemisphere. The present data thus suggest formation of foil-excited atoms by backward capture processes. We suspect there are useful analogies to be drawn with the theory[8] of continuum electron capture in binary collisions. The data at the various field strengths and energies are being analyzed in more detail and will be reported elsewhere.

We are extremely grateful for the insights into interpretation of this data provided by T. G. Eck, who also stimulated this work. We thank B. R. Appleton and O. E. Schow of the Oak Ridge National Laboratory solid state division for providing accelerator facilities and invaluable technical help in these experiments. We should also like to acknowledge T. A. Welton's original suggestion[9] that backward capture processes might predominate in these collisions.

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