Nonadiabatic Behavior of the Polarization of Electric-Field-Induced Lyman-α Radiation

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The linear polarization of Lyman-α radiation emitted when hydrogen atoms in the metastable $2s_{1/2}$ state traverse an external electric field is studied as a function of the effective rise time of the field for field strengths extending from very small values up to 6 keV/cm. With decreasing rise time, the measured polarization exhibits a transition from *adiabatic* to *sudden* behavior. The transition region is centered at a rise time which is approximately equal to the characteristic time associated with the $2s_{1/2}$-$2p_{1/2}$ Lamb splitting in the unperturbed hydrogen system. The experimental data are well described by the results of a time-dependent theory.

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When a beam of hydrogen atoms prepared in the metastable $2s_{1/2}$ state traverses an external electric field, the Stark mixing among the $2s_{1/2}$, $2p_{1/2}$, and $2p_{3/2}$ states induces the emission of Lyman-α radiation. The linear polarization P as well as the closely related anisotropy R of this radiation have been investigated both experimentally and theoretically in a number of cases.[1-14] The emphasis in these studies has been on the low-field regime, with the field strength F not exceeding values of a few hundred V/cm. Particular interest has been devoted to the limit $F \rightarrow 0$ and the possibility of deriving the $2s_{1/2}$ Lamb shift by measuring P or R in this limit.[9-13] A first study extending into the high-field regime ($F \leq 6$ kV/cm) has been performed only recently by Harbich *et al.*[14]

The conditions of the previous experimental investigations were characterized by a *slow* entrance of the metastable hydrogen atoms into the field region. More quantitatively, the effective rise time τ of the field as seen by the atoms in their rest frame was large as compared with the characteristic times associated with the $2p_{3/2}$-$2p_{1/2}$ fine-structure splitting and the $2s_{1/2}$-$2p_{1/2}$ Lamb splitting in the unperturbed hydrogen system. In terms of the "adiabaticity" parameter $\eta = T/\tau$, where T is the relevant characteristic time in the unperturbed system, the adiabatic limit $\tau \rightarrow \infty$ corresponds to $\eta = 0$, and the experiments performed so far are characterized by $\eta \ll 1$. This near adiabaticity is markedly expressed in the measured polarization of Harbich *et al.*,[14] which is in close agreement with theoretical results obtained within a strictly adiabatic approximation, in particular at large field strengths where it approaches the theoretical value $P = -100\%$ calculated in the limit $F \rightarrow \infty$.

In this Letter, we present and discuss results of an experimental and theoretical investigation in which the polarization of field-induced Lyman-α radiation is for the first time systematically studied in the *nonadiabatic* regime ($\eta \simeq 1$ and $\eta \gg 1$) at both small and large field strengths. These results are believed to be of general interest as they contribute to the detailed understanding of a prototype case of polarization of atomic line radiation. Specifically, the present study attempts to answer the question[8] as to which one of the two characteristic times associated with the hydrogen $n = 2$ system, that related to fine structure or the one related to the Lamb splitting, is the relevant time that controls the nonadiabatic behavior of the polarization of field-induced Lyman-α radiation.

Figure 1 shows a schematic view of our experimental setup, which is similar to that used in Ref. 14. In brief, protons with kinetic energies between 5 and 20 keV are extracted from a duoplasmatron ion source and are neutralized in an argon-filled charge-exchange cell. The hydrogen beam (diameter ≈ 2.0 mm), after having traveled a sufficiently large distance over which excited states (except the metastable $2s_{1/2}$ state) have decayed to the ground state, enters a *longitudinal* electric field ($0 \leq F \leq 6$ kV/cm) through the circular aperture in the front plate of a parallel-plate capacitor (plate spacing,

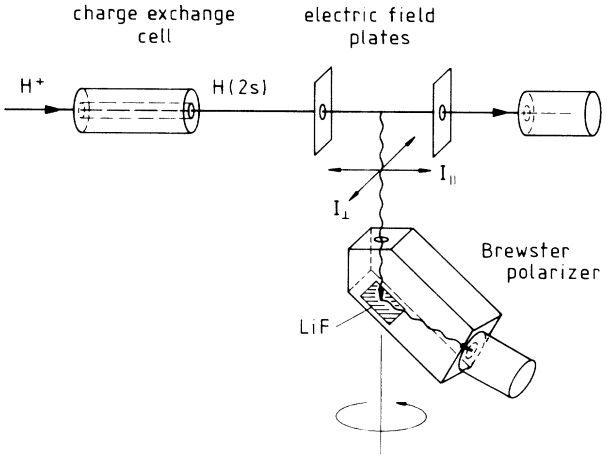

FIG. 1. Schematic view of the experimental setup (not to scale). The arrows marked $I_{\parallel}$ and $I_{\perp}$ refer to the intensities of the radiation with polarization parallel and perpendicular to the field direction, respectively [cf. Eq. (6)].

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10.16 ± 0.05 mm; plate thickness, 0.635 ± 0.025 mm). By exchanging the front plate, the diameter D of the aperture can be varied between 0.1 and 3.0 mm. The field-induced Lyman-α radiation is observed at 90° with respect to the hydrogen beam. The linear polarization of this radiation is measured by means of a Brewster-angle polarizer equipped with a LiF plate.

The field distribution $\mathcal{F}(z)$ along the symmetry axis of the aperture, obtained from a numerical calculation[15] taking into account the finite thickness of the capacitor plates, is shown in Fig. 2 for various D values. The effective rise time τ (which will be defined quantitatively below) depends, in good approximation, *linearly* on the ratio D/v, where v is the beam velocity. By varying v and/or D, we are able to vary τ roughly within the range $10^{-11}\ \mathrm{s} \leq \tau \leq 10^{-9}\ \mathrm{s}$.

Before turning to our experimental results, we outline the theoretical framework within which these results will be analyzed. In the rest frame of the hydrogen atoms (with the z axis along the field direction), the atom-field coupling is described by the time-dependent Hamiltonian (unless stated otherwise, we use atomic units)

$$H(t) = H_0 + \mathcal{F}(t)z - i\Gamma/2 \equiv H_1(t) - i\Gamma/2 , \quad (1)$$

where H_0 denotes the unperturbed atomic Hamiltonian which includes fine-structure effects within the Pauli approximation[16] as well as a phenomenological correction accounting for the $2s_{1/2}$ Lamb shift[17] (hyperfine structure is neglected; its influence will be discussed below). The field-strength function $\mathcal{F}(t)$ [with $\mathcal{F}(t=-\infty)=0$ and $\mathcal{F}(t=\infty)=F$] is obtained from the field distribution $\mathcal{F}(z)$ in the laboratory frame by means of the transformation $z=vt$. The "decay operator" Γ is diagonal in the unperturbed atomic representation, with the nonzero diagonal elements given by the radiative decay widths of the $2p_{3/2}$ and $2p_{1/2}$ states. The Hamiltonian $H(t)$ commutes with the projection of the total electronic angular momentum along the field direction, so that the quantum number m_j is a good quantum number.

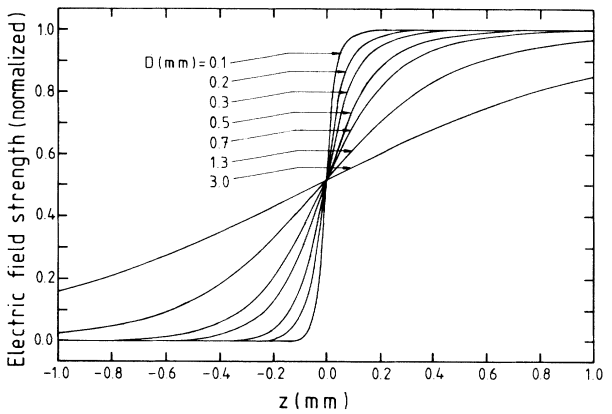

FIG. 2. Numerically calculated field distribution $\mathcal{F}(z)$ along the symmetry axis of the aperture, normalized to the value $F=\mathcal{F}(z=\infty)$, for various values of the diameter D. The point $z=0$ corresponds to the inner boundary of the aperture.

In order to solve the time-dependent Schrödinger equation $[H(t) - i\partial/\partial t]\Psi(t) = 0$, we expand the wave function $\Psi(t)$ in terms of adiabatic states $\psi_j^{\mathrm{ad}}(t)$ ("Stark states") obtained by diagonalizing, at fixed time t, the Hamiltonian $H_1(t)$ in the space of the unperturbed $2p_{3/2}$, $2s_{1/2}$, and $2p_{1/2}$ states (each corresponding to $m_j = \frac{1}{2}$):

$$H_1(t)\psi_j^{\mathrm{ad}}(t) = \epsilon_j^{\mathrm{ad}}(t)\psi_j^{\mathrm{ad}}(t), \quad j=1,2,3 , \quad (2)$$

$$\Psi(t) = \sum_{j=1}^{3} a_j(t)\psi_j^{\mathrm{ad}}(t)\exp\left[-i\int_{-\infty}^{t} dt'\,\epsilon_j^{\mathrm{ad}}(t')\right] . \quad (3)$$

The amplitudes $a_j(t)$ are determined from the system of coupled equations

$$i\dot{a}_j(t) = \sum_{j'=1}^{3} M_{jj'}(t)a_{j'}(t) , \quad (4)$$

with the coupling matrix elements $M_{jj'}$ given by

$$M_{jj'}(t) = -i\langle\psi_j^{\mathrm{ad}}(t)|\partial/\partial t + \Gamma/2|\psi_{j'}^{\mathrm{ad}}(t)\rangle \times\exp\left[-i\int_{-\infty}^{t} dt'[\epsilon_j^{\mathrm{ad}}(t') - \epsilon_{j'}^{\mathrm{ad}}(t')]\right] . \quad (5)$$

The initial conditions are $|a_{2s_{1/2}}(t=-\infty)|=1$, $a_{2p_{3/2}}(t=-\infty)=a_{2p_{1/2}}(t=-\infty)=0$. The matrix elements of the $\partial/\partial t$ operator in Eq. (5) are easily evaluated by means of the Hellmann-Feynman theorem.

It is noted that our theoretical approach is similar to that employed by Drake[18] in a study of field-induced quantum beats in hydrogen. The main difference is that Drake uses a time-independent basis defined by the eigenfunctions of the full (complex) Hamiltonian $H(t=\infty)$ corresponding to the maximum field strength F, while we use the (time-dependent) adiabatic basis defined by the eigenfunctions of the real Hamiltonian $H_1(t)$.

The intensities $I_{\parallel}(t)$ and $I_{\perp}(t)$ of Lyman-α radiation emitted at time t with polarization parallel and perpendicular to the field direction, respectively, are readily expressed in terms of the dipole matrix elements connecting the states $\psi_j^{\mathrm{ad}}(t)$ with the $1s_{1/2}$ ground state and of the density matrix constructed from the amplitudes $a_j(t)$. The linear polarization P is given by

$$P = (\bar{I}_{\parallel} - \bar{I}_{\perp})/(\bar{I}_{\parallel} + \bar{I}_{\perp}) , \quad (6)$$

where the intensities $\bar{I}_{\parallel}$ and $\bar{I}_{\perp}$ are obtained by integrating $I_{\parallel}(t)$ and $I_{\perp}(t)$ over the time interval corresponding to the "observation window" of the photon detector (which extends from 1.6 to 5.4 mm behind the aperture).

A selection of our results is displayed in Fig. 3, where the linear polarization of the field-induced Lyman-α radiation is plotted as a function of the field strength F for various values of the diameter D and for a kinetic energy of the hydrogen atoms of 20 keV. The theoretical curves have been obtained by using numerical field-strength

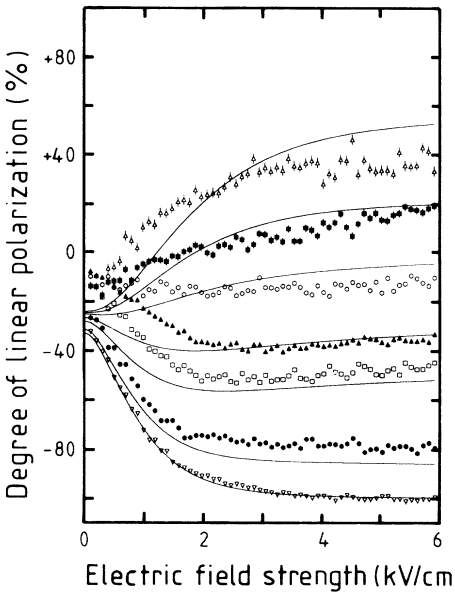

FIG. 3. Linear polarization of field-induced Lyman-α radiation at a kinetic energy of the hydrogen atoms of 20 keV, plotted as a function of the field strength F for a diameter D of the entrance aperture of 0.1 mm (△), 0.2 mm (■), 0.3 mm (○), 0.5 mm (▲), 0.7 mm (□), 1.3 mm (●), and 3.0 mm (▽), respectively. The solid curves refer to the theoretical calculations as explained in the text.

functions $\mathcal{F}(t)$ corresponding to the curves of Fig. 2. Corrections taking into account the inhomogeneity of the field over the aperture have not been applied.

A general trend observed in the data of Fig. 3 is the increase of the polarization with decreasing diameter D, i.e., with decreasing rise time τ. This trend is to be qualitatively expected since, for sufficiently small τ and sufficiently large field strength F, the polarization should approach[14] a value of $+100\%$.

The agreement between experimental and theoretical results in Fig. 3 is, in general, quite good. Marked deviations are found only in the range $F \leq 2$ kV/cm and $D \leq 0.7$ mm. We are not able to offer a complete explanation for these discrepancies. Effects of hyperfine structure, which are neglected in the present calculations, are expected to be small. A previous calculation[3] had shown that the (absolute) correction to the polarization due to hyperfine structure is about 0.5% in the adiabatic limit at very small F. An even smaller correction is anticipated for larger F and finite rise times. It is noted that the experimental data for $D \leq 0.7$ mm appear to converge towards a common value of P in the limit $F \rightarrow 0$. This value is close to the value $P = -14.7\%$ obtained for $F \rightarrow 0$ in the strict *sudden* limit ($\eta = \infty$). A partial explanation for this finding is provided by the fact that for small D the beam profile completely covers the entrance aperture. In this case, the hydrogen atoms traveling close to the boundary of the aperture experience a field that rises much more steeply than that on the symmetry axis (which is the field assumed in the theoretical calculation). However, if the inhomogeneity of the field over the beam profile would be the only reason for the discrepancies, the measured polarization should be larger than the theoretical values for all F values. Obviously, the incomplete knowledge of the function $\mathcal{F}(t)$ (due to imprecisely known boundary conditions) as well as uncertainties in the location of the observation window constitute further sources for discrepancies between experiment and theory.

The data shown in Fig. 3 do not provide any evidence for the presence of beatlike structure in the field dependence of the polarization. The theoretical polarization exhibits rapid oscillations when calculated from the intensities at a fixed position within the field. These oscillations which are associated with the nondiagonal elements of the density matrix ("coherences") are completely smeared out by the integration over the observation window. Under the present conditions, the nondiagonal elements may in fact be safely omitted in the calculations.

Results similar to those shown in Fig. 3 for 20 keV have been obtained at beam energies of 5, 10, and 15 keV. A similar overall trend of the data and of the discrepancies between experiment and theory is observed in these cases.

For a unified discussion of our results, we display in Fig. 4 the measured polarization for selected values of F as a function of the reciprocal of the effective rise time τ, i.e., on a scale that is proportional to the adiabaticity parameter η. While no unambiguous definition of τ exists, it seems appropriate to identify this quantity with the full width at half maximum of the function $d\mathcal{F}/dt$, which is a bell-shaped function centered at the time corresponding to the position of the entrance aperture. In Fig. 4, we use τ values defined as $\tau = 0.4238\tau_0$, where τ_0 is the value obtained from the *numerically* calculated function $\mathcal{F}(t)$ on the symmetry axis. Within less than 10%, τ_0 agrees with the value $0.6436D/v$ derived from an *analytic* solution for $\mathcal{F}(t)$ for zero thickness of the capacitor plates.[19] The correction factor 0.4238, which takes into account the inhomogeneity of the field over the aperture (assuming a homogeneous beam profile), has been evaluated from the analytic solution for $\mathcal{F}(t)$.

The measured polarization is seen to depart from the calculated adiabatic values for τ less than about 0.3×10^{-9} s and to approach the calculated sudden values monotonically with decreasing τ. The center of the transition region from adiabatic to sudden behavior, which may be defined as the position of the steepest rise of the polarization, is located approximately at $\tau = 1 \times 10^{-10}$ s. This value is close to the characteristic time $T_L = 1.505 \times 10^{-10}$ s associated with the $2s_{1/2}$-$2p_{1/2}$ Lamb splitting in the unperturbed hydrogen system (we define T_L in the usual way via the energy-time uncer-

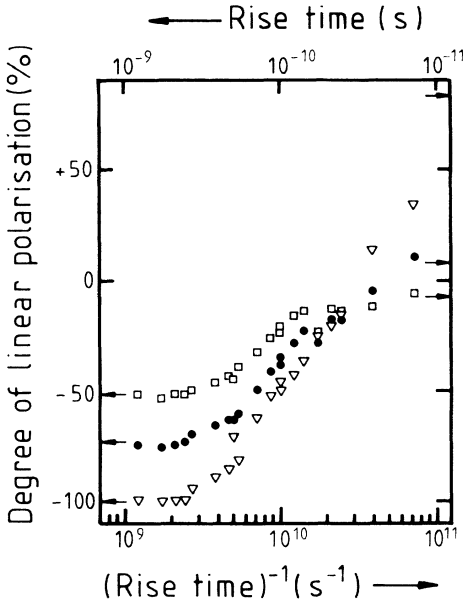

FIG. 4. Measured linear polarization of field-induced Lyman-α radiation, plotted as a function of the reciprocal of the effective rise time τ for a field strength F of 0.5 kV/cm (□), 1.0 kV/cm (●), and 5.0 kV/cm (▽), respectively. For the definition of τ, see text. The arrows attached to the left-hand and right-hand ordinates indicate (in correspondence with the order of the nearest experimental points) the *theoretical* polarization calculated in the *adiabatic* approximation and in the *sudden* approximation, respectively.

tainty relation; this definition differs from that of Ref. 8 by a factor of 2π). The characteristic time T_{FS} associated with the $2p_{3/2}$-$2p_{1/2}$ fine-structure splitting is smaller than T_L by about 1 order of magnitude.

In conclusion, nonadiabatic behavior of the linear polarization of electric-field-induced Lyman-α radiation at both small and large field strengths has been exhibited for the first time. The transition region from adiabatic to sudden behavior is centered at a rise time of the field which is close to the characteristic time associated with the $2s_{1/2}$-$2p_{1/2}$ Lamb splitting. The measured polarization in the nonadiabatic regime was found to be in good agreement with the results of a time-dependent theory. Further insight into the role of the different interactions in the hydrogen $n=2$ system and the characteristic times associated with them can undoubtedly be gained from a more detailed theoretical analysis.

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