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Citation: *Appl. Phys. Lett.* **69**, 3749 (1996); doi: 10.1063/1.117180

View online: <http://dx.doi.org/10.1063/1.117180>

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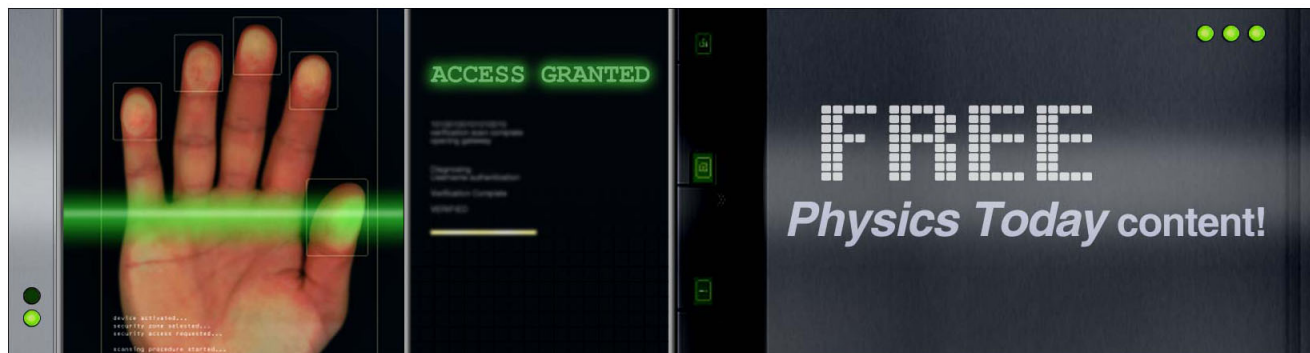
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Measurement of rapidly varying electric fields through parity oscillations in the Rydberg states of hydrogenic atoms

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(Received 19 August 1996; accepted for publication 1 October 1996)

Oscillations are shown to exist in the inversion symmetry of the electronic wave function of a hydrogenic atom coherently excited to a Rydberg state by a short pulse of laser radiation in a uniform electric field. The dependence of these oscillations on field strength is shown to scale as n^2 where n is the principal quantum number. The possibility of using these oscillations to measure electric signals on picosecond timescales (terahertz frequencies) is suggested. © 1996 American Institute of Physics. [S0003-6951(96)00650-X]

The promise of new ultrafast electric and optoelectronic devices has driven the development of techniques to detect electromagnetic radiation on picosecond and subpicosecond timescales.¹⁻⁷ One detection strategy has been to use a laser-gated detector to probe a pulse of terahertz radiation created by a laser-driven emitter.¹⁻⁶ A second strategy has been to measure the interference between pulses of radiation created by two laser-driven emitters.⁷ In both cases the autocorrelation between two short-pulse laser-drive processes is measured. These pump-probe detection schemes are somewhat limited. As a probe of the properties of an optoelectronic emitter, for example, these schemes require that the emitter produce a repeatable pulse profile. As an instrument for communication, these schemes require precise synchronization between the laser-driven transmitter and the laser-gated receiver.

These limitations could be overcome by replacing a single spatially fixed detector with a continuous series of detectors that sample the terahertz radiation as it passes without distorting it significantly. The ability to image the profile of single pulse of radiation would both allow for the characterization of transient events and simplify the technological challenge of terahertz communication. In this letter we propose that the Rydberg states of hydrogenic atoms can be used to create detectors of electric field strength that may be ideally suited for such measurements.

The exploitation of the sensitivity of highly excited Rydberg states of atoms to their surroundings is not a new idea. In 1976 Kleppner and Ducas⁸ proposed that the excited states of hydrogen could be used to detect both microwave and near infrared radiation. This realization led to the development of sensitive near infrared detectors.⁸⁻¹⁴ It is shown here that the Stark mixing of the angular momentum states of highly excited hydrogenic atoms leads to oscillations in the inversion symmetry of the electronic wave function. The electric field surrounding a hydrogenic atom can be measured on a picosecond timescale by probing these oscillations with short pulses of laser radiation.

In the Stark effect, an electric field splits the otherwise degenerate states of hydrogen into states of energy $-Z^2/(2n^2) + [3nF/(2Z)]k$. Here Z is the charge of the nucleus, n is the principal quantum number, F is the field

strength, and k is an electric quantum number that is related to the parabolic quantum numbers¹⁵ n_1 and n_2 by $k = n_1 - n_2$. Unless otherwise specified, all quantities in this letter will be expressed in atomic units. One way to observe the Stark effect is to observe the quantum beats exhibited by an atom excited by a short pulse of laser radiation.¹⁶ If the bandwidth of the laser radiation is much broader than the $3n(n-1)F/Z$ width of the Stark manifold, the electron is excited to a Rydberg state that is initially indistinguishable from the Rydberg state excited in the absence of the applied electric field. The initial wave function of the electron is not, however, a steady-state solution to the Schrödinger equation. For this reason the wave function evolves with time. If the initial state of the atom is described by the quantum numbers n , l , and $m=0$, the time-dependent wave function of the electron will be given by

$$\psi_{nl0}(\mathbf{r}, t) = \sum_{k=1-n, 3-n, 5-n}^{n-1} \langle nk0 | n l 0 \rangle | nk0 \rangle e^{i\omega k t}. \quad (1)$$

Here $|nk0\rangle$ are the eigenfunctions of the perturbed Hamiltonian with eigenvalues ωk where $\omega = 3Fn/2Z$.

The time-dependent expectation value of the parity operator, $\langle \Pi(t) \rangle = \langle \psi_{nl0}(\mathbf{r}, t) | \psi_{nl0}(-\mathbf{r}, t) \rangle$ may be evaluated using Eqs. (1) and the solution for the spherical-parabolic matrix elements $\langle nlm | nkm \rangle$ determined by Park:¹⁷

$$\langle \Pi(t) \rangle = (-1)^l (2l+1) \times \sum_{k=1-n, 3-n, 5-n}^{n-1} \begin{pmatrix} \frac{n-1}{2} & \frac{n-1}{2} & l \\ k & -k & 0 \end{pmatrix}^2 e^{-2i\omega k t}. \quad (2)$$

Here the bracketed term in the summation is a $3-j$ symbol. For an electron initially excited to a $p(l=1)$ state Eq. (2) reduces to

$$\langle \Pi(t) \rangle = \frac{3}{(n^2-1)n} \frac{1}{4\omega^2} \frac{d^2}{dt^2} \left[\frac{\sin(2n\omega t)}{\sin(2\omega t)} \right]. \quad (3)$$

The time-dependent parity exhibits rapid oscillations indicated by the $\sin(2n\omega t)$ numerator of the bracketed term in Eq. (3). These high-frequency oscillations in the parity of the excited state occur at a frequency of $\omega_p = 2n\omega = 3n^2F/Z$ and appear to be purely quantum mechanical in nature. The fact that ω_p increases as n^2 implies that, for large n , the frequency of the parity oscillations, and hence the evolution of the Rydberg state of an atom, is extremely sensitive to the strength of an applied electric field.

To observe parity oscillations, the selection rule that requires the parity of the electronic state to change in a dipole transition may be exploited. In one of many possible schemes, hydrogenic atoms are excited to a Rydberg state via the $1s \rightarrow 50p$ transition with linearly polarized light. The excited hydrogen atoms are then allowed to evolve in a weak electric field that is parallel to the laser polarization for a time delay of 50–1000 ps. The time-dependent shape of the evolving wave function is then probed by inducing the $n=50 \rightarrow n=2$ transition and observing the subsequent Lyman alpha fluorescence. We emphasize that the 50–1000 ps time delay is between the laser radiation that excites ground state hydrogen atoms to the $50p$ state and the laser radiation that induces the $n=50 \rightarrow n=2$ transition. The subsequent Lyman alpha fluorescence gives the efficiency that this transition is induced. This fluorescence will have a bimodal decay with a rapid component corresponding to the fluorescence of the $2p$ state and a slow component corresponding to the fluorescence of the $2s$ state. Here, for simplicity, it is assumed that only the slow $2s$ -state fluorescence is observed.

The ratio of the time-dependent- to the time-independent-matrix element of the $50p \rightarrow 2s$ transition is given by

$$I_n(t) = \frac{|\langle 210|z|\psi_{n10}(t)\rangle|^2}{|\langle 200|z|n10\rangle|^2} = \left| \sum_{k=1-n,3-n,5-n}^{n-1} |\langle nk0|n10\rangle|^2 e^{i\omega_k t} \right|^2, \quad (4)$$

which reduces to

$$I_n(t) = \left| \frac{3}{(n^2-1)n^2\omega^2} \frac{d^2}{dt^2} \left[\frac{\sin(n\omega t)}{\sin(\omega t)} \right] \right|^2. \quad (5)$$

A plot of $I_n(t)$ versus time shows low-frequency oscillations which are the result of the fact that the electric field pulls the otherwise stationary $50p$ orbital from a region close to the nucleus to a region far removed from the nucleus and back again (Fig. 1). The period of these oscillations is given by $\tau_0 = 2\pi/3$ nF; it is equal to 5.210 ns for $F = 1$ V cm⁻¹ and $n = 50$. Recurrences of this type have been observed by ten Wolde *et al.*,¹⁸ who reported that the ionization probability of the $n=23$ Rydberg state of rubidium in a field of 247.5 V cm⁻¹ recurs with the predicted period of $\tau_0 = 2\pi/3$ nF = 45.8 ps.

The analysis presented here predicts additional oscillations in the expected efficiency for induced emission at the frequency of the parity oscillations [Fig. 1 (inset)]. Unlike the lower-frequency oscillations at the Stark splitting, these oscillations are not caused by motion of the electronic probability from a region near the nucleus to a region far from the

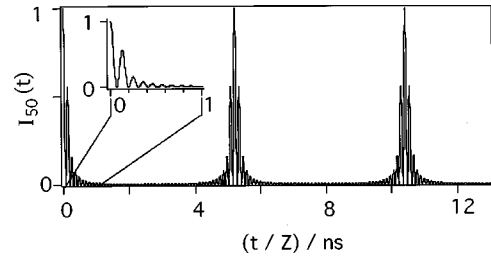


FIG. 1. The time-dependent signal expected from a pump-probe study of the evolution of hydrogen atoms excited to a $50p$ state in the presence of a 1 V cm⁻¹ electric field. Here $I_{50}(t)$ is the ratio of the time-dependent oscillator strength of the $n=50$ to $2s$ transition to the time-independent oscillator strength of the $50p$ to $2s$ transition.

nucleus. Instead, these oscillations reflect the parity oscillations of the Rydberg orbital caused by field-induced l mixing. Oscillations of this nature have yet to be observed experimentally. The fact that a field strength of only 1 V cm⁻¹ induces a frequency-dependent signal on the order of 10^4 ps is remarkable. This result implies that an excited hydrogen atom could be used to sample weak electrical signals on a picosecond timescale.

The measurement described assumes that the bandwidth of the excitation radiation is broad enough to excite an entire Stark manifold but narrow enough not to excite adjacent Stark manifolds. These conditions cannot be met if too short a pulse of laser radiation is employed. A pulse of laser radiation with a Gaussian time-dependent intensity profile characterized by Δt_{FWHM} has an angular frequency resolution of $\Delta\omega_{\text{FWHM}} = 4\ln 2/\Delta t_{\text{FWHM}}$ [FWHM (full width at half-maximum)]. This condition places a limit on the minimum pulse width of an excitation laser that is used to excite a single n state. Given the spacing between adjacent n levels in a hydrogen atom is $1/n^3$, excitation of only one n level requires Δt_{FWHM} to be much greater than $4n^3\ln 2$. For $n=50$ we find this condition implies that Δt_{FWHM} must be greater than 8 ps.

To explore the possibility of faster measurements of electric field strength, we predict the field-dependent efficiency $I(F, \Delta t_{\text{FWHM}})$ with which two coincident pulses of laser radiation populate the $2s$ state of a hydrogenic atom (Fig. 2). The center frequency of one of these pulses is tuned

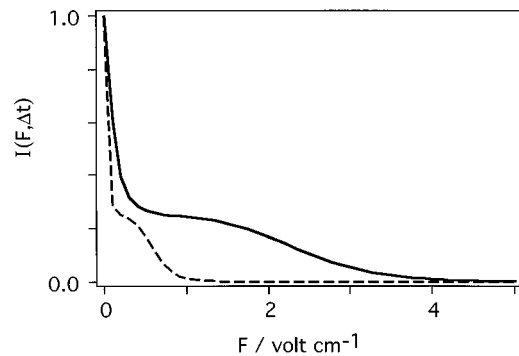


FIG. 2. Dependence of the efficiency for excitation to the $2s$ state of a hydrogenic atom via the induced transitions $1s \rightarrow H(\text{high } n) \rightarrow 2s$ on electric field strength for $\Delta t_{\text{FWHM}} = 5$ ps (solid line) and $\Delta t_{\text{FWHM}} = 10$ ps (dashed line). Each curve is normalized to the efficiency for excitation to the $2s$ state under field-free conditions.

to the field-free ionization limit for the $1s \rightarrow H^+$ transition whereas the second frequency is tuned to the field-free ionization limit for the $2s \rightarrow H^+$ transition. Because of the inherently broad bandwidth of the short pulse of radiation tuned to the $1s \rightarrow H^+$ transition, a spectrum of high n states will be excited. Similarly, the short pulse of laser radiation tuned to the $2s \rightarrow H^+$ transition will drive many of these electronic states back to the $2s$ state. To obtain the result shown in Fig. 2, the analysis leading to the result of Eq. (5) was modified to include averaging over both the distribution of n states within the bandwidth of the laser radiation, and over the uncertainty in the time delay between laser-induced excitation and laser-induced emission. Details of this calculation will be given in a future work.

One interesting feature of $I(F, \Delta t_{\text{FWHM}})$ is the broad shoulder of the sharp peak at zero field strength. This feature occurs as the increasing field shifts low- n quantum states into the bandwidth of the laser radiation. A second observation is the strong dependence of $I(F, \Delta t_{\text{FWHM}})$ on the duration of the laser pulse. This dependence demonstrates the inherent trade-off between sensitivity to electric field and time resolution of the measurement.

This discussion has been limited to hydrogenic systems. The analysis of multielectron atoms and molecules requires consideration of the influence of quantum defects and the competition between diabatic and adiabatic motion of the electron. We recognize that the difficulty of manipulating hydrogen atoms and creating vacuum ultraviolet light to excite them may make a method of measuring electric field strength based on the detection of nonhydrogenic systems more practical. The simple analysis presented here, however,

demonstrates the rapid timescale for l mixing and suggests a possible use of the Rydberg states of atoms and molecules as a probe of high frequency electrical signals.

This work is supported in part by funding from the University of Oklahoma and from the National Science Foundation under NSF Grant No. CHE-9322690.

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