Rydberg wave packets in parallel electric and magnetic fields

I. Moser, F. Mota-Furtado, P. F. O'Mahony

Department of Mathematics, Royal Holloway, University of London, Egham, Surrey TW20 OEX, United Kingdom

J. P. dos Santos

*Centro de Física Atómica da Universidade de Lisboa and Departamento de Física da Universidade Nova de Lisboa,
Avenida Prof. Gama Pinto 2, P-1699 Lisboa Codex, Portugal*

The magnitude of the time autocorrelation function M between states excited by two Gaussian laser pulses is calculated for both hydrogen and rubidium atoms in parallel electric and magnetic fields. M is determined by a full quantum-mechanical calculation but the peaks are identified with the periods of the shortest periodic orbits of the corresponding classical system. Qualitative agreement is obtained with experimental results, however, discrepancies are found in the relative heights of the peaks. [S1050-2947(97)01105-0]

PACS number(s): 32.80.Rm, 32.60.+i

I. INTRODUCTION

The frequencies or periods associated with classical periodic orbits play an important role in interpreting the spectra of atoms in external fields. These frequencies are in particular associated with unstable periodic orbits in the classically chaotic region of the spectrum. Until recently experiments have identified these periodicities indirectly by measuring the absorption spectrum over a wide energy range and then taking a Fourier transform of this frequency domain spectrum [1,2]. Over the last few years, new experiments have attempted to identify these periodicities directly in the time domain [3–9]. This is done by using short pulsed lasers to create Rydberg wave packets whose return times to the vicinity of the nucleus or core are measured, giving directly the periods of the classical orbits. The initial Rydberg wave-packet experiments used a pump-probe technique where two laser pulses separated by a time delay t_d [10,3] are incident on an atom. The first pulse (the pump) excites a wave packet from a low-lying state and the second pulse (the probe) attempts to ionize the wave packet. The photoionization process only occurs if the Rydberg electron is near the core of the atom because the nucleus is required to act as a third body in order to conserve both energy and momentum. Peaks in the number of ionized electrons as a function of t_d therefore give a measurement of the return time of the wave packet to the core region. The major problem with this method is that experimentally it produces a poor signal-to-noise ratio.

This drawback prompted a method based on a phase sensitive technique [11,12]. This method uses a similar scheme of two laser pulses separated by a short time delay, but now it relies on interference effects between two wave packets excited by two identical laser pulses as opposed to the photoionization process. As before the first pulse excites a wave packet, but now the second pulse creates an identical wave packet, which can interfere with the time evolved initial wave packet. If the wave packet excited by the first pulse is far from the core of the atom when the second wave packet is excited there is no overlap between the two wave packets and no interference will occur. If, however, the initial wave

packet has returned to near the core, the second wave packet will interfere with the first in such a way that the Rydberg population is either enhanced or depleted. The final Rydberg population, after the pulses have interacted with the atom, is measured by field ionization. This is a very efficient measurement process giving a much better signal-to-noise ratio than the pump-probe technique. The quantity that can be extracted from the experiment is the magnitude of the autocorrelation function $M = |\langle \Psi(0)|\Psi(t_d)\rangle|$, which measures the overlap between the initial wave packet, $|\Psi(0)\rangle$, and the wave packet at some time t_d later. This function plays an important role in the semiclassical theory of chaos [13].

Marmet *et al.* [4] have used the above technique to measure M as a function of t_d for the rubidium atom in both magnetic and parallel electric and magnetic fields. They have also evaluated M semiclassically. The semiclassical theory, which was carried out for hydrogen, failed to agree with the experimental results for parallel fields and it was stated that this may be due to the neglect of the core potentials of rubidium.

In this paper we present a calculation of the autocorrelation function M as a function of t_d for an atom in parallel electric and magnetic fields subject to two Gaussian laser pulses. As in the experiment, the field strengths and energies are chosen in a regime where one observes a transition from regular to irregular behavior in the spectrum corresponding to a classical transition from regularity to chaos. M is evaluated via an *ab initio* quantum-mechanical calculation for both hydrogen and rubidium in parallel fields allowing a direct examination of nonhydrogenic core effects. The peaks in M are compared with orbits whose periods have been determined from classical calculations. Qualitative agreement between theory and experiment for the position of peaks is obtained for rubidium, however, quantitative agreement for the heights of the peaks is still lacking.

In Sec. II we give a brief description of the experimental determination of M, the perturbative derivation of an expression for M and its evaluation for Rydberg atoms in parallel fields. In Sec. III we compare the theoretical results for hydrogen and rubidium with experiment. Atomic units are used throughout unless otherwise stated.

II. THEORY

A. The autocorrelation function

An atom is excited from its ground state by a pair of identical laser pulses separated by a time t_d. The first pulse, at time $t=0$, creates a Rydberg wave packet represented by $|\Psi(0)\rangle$, which can be expanded as a superposition of eigenstates of the atom. The coefficients of this superposition depend on both the form of the pulse and the dipole moments characterizing transitions from the ground state to the relevant Rydberg states. The state $|\Psi(0)\rangle$ evolves for a time t_d at which time a second laser pulse excites an identical Rydberg wave packet. The total wave function is therefore $|\Psi_{\mathrm{Ryd}}\rangle = |\Psi(0)\rangle + |\Psi(t_d)\rangle$. The total Rydberg population P_{tot} is proportional to $\langle\Psi_{\mathrm{Ryd}}|\Psi_{\mathrm{Ryd}}\rangle$ or

$$P_{\mathrm{tot}} \propto \langle\Psi(0)|\Psi(0)\rangle + |\langle\Psi(0)|\Psi(t_d)\rangle| \cos\chi(t_d), \quad (1)$$

where χ denotes the phase difference between the wave packets excited by the first and second pulses. Clearly M will be large whenever the overlap between the first and second wave packets is large. The total Rydberg population has in general a rapid sinusoidal time dependence around an average value, the amplitude of which is proportional to the magnitude of the autocorrelation function M. By using an additional phase delay M can be extracted from the experiments and measured by field ionization of the Rydberg states [7,4].

B. The autocorrelation function for an atom excited by two Gaussian pulses

When the intensity of the laser pulses is low, as in the experiment of Marmet *et al.* [4], the interaction potential between the laser field and the atom, $V(t)$, can be treated by time-dependent perturbation theory. The total Hamiltonian for the system is then written as

$$H = H_0 + V(t), \quad (2)$$

where H_0 is the time-independent Hamiltonian for the atom in the presence of the static electric and magnetic fields. The ground state of H_0 will be denoted by $|g\rangle$ and the excited eigenstates by $|j\rangle$. The potential $V(t) = -\boldsymbol{\mu}\cdot\mathbf{E}(t)$ where $\boldsymbol{\mu}$ is the electric dipole vector and $\mathbf{E}(t)$ the classical electric field due to the laser.

We take the atom, in the presence of the static fields, to be initially in its ground state $|g\rangle$. Expanding the total wave function in terms of the unperturbed states, the time-dependent wave function $|\Psi\rangle$ is given by

$$|\Psi\rangle = |g\rangle a_g(t) e^{-i\omega_g t} + \sum_j |j\rangle a_j(t) e^{-i\omega_j t}, \quad (3)$$

where the sum is taken over all excited states and $a_g(t), a_j(t)$ represent the time-dependent amplitudes of the ground and excited states with energies ω_g, ω_j. Note that the sum over $|j\rangle$ must contain a sum over all the relevant quantum numbers.

The time-dependent electric field $\mathbf{E}(t)$ for the excitation scheme of two Gaussian pulses of pulse width τ and angular frequency ω_l is given by

$$\mathbf{E}(t) = \mathbf{E}_0 e^{i\omega_l t} e^{-2\ln 2(t/\tau)^2} + \mathbf{E}_0 e^{i\omega_l(t-t_d)} e^{-2\ln 2[(t-t_d)/\tau]^2}, \quad (4)$$

hence the time-dependent potential $V(t)$ is known. By substituting $V(t)$ and the expansion in Eq. (3) into the time-dependent Schrödinger equation, a standard set of equations for the time-dependent Rydberg amplitudes is obtained. Using the rotating-wave approximation and neglecting depletion of the ground state [14], the excited-state amplitudes in first order in perturbation theory are given by

$$a_j(t) = \langle j|\boldsymbol{\mu}\cdot\mathbf{E}_0|g\rangle \int_{-\infty}^t e^{i(\omega_j - \omega_g - \omega_l)t'} \big(e^{-2\ln 2(t'/\tau)^2} + e^{-2\ln 2[(t'-t_d)/\tau]^2} e^{i\omega_l t_d} \big) dt'. \quad (5)$$

Taking the upper limit of the integral as ∞ (measurement of the Rydberg population occurs long after the interaction of the pulses) yields the coefficients

$$a_j = \sqrt{\frac{\pi}{2\ln 2}} \tau \langle j|\boldsymbol{\mu}\cdot\mathbf{E}_0|g\rangle e^{-[(\omega_j - \omega_g - \omega_l)^2/8\ln 2]\tau^2} \times (1 + e^{i(\omega_j - \omega_g)t_d}). \quad (6)$$

The wave function of the excited or Rydberg wave packet is therefore

$$|\Psi_{\mathrm{Ryd}}\rangle = \sqrt{\frac{\pi}{2\ln 2}} \tau \sum_j e^{-i\omega_j t} |j\rangle\langle j|\boldsymbol{\mu}\cdot\mathbf{E}_0|g\rangle \times e^{-[(\omega_j - \omega_g - \omega_l)^2/8\ln 2]\tau^2} (1 + e^{i(\omega_j - \omega_g)t_d}). \quad (7)$$

The total Rydberg wave function can be written as the sum of the first and second excited wave packets,

$$|\Psi_{\mathrm{Ryd}}\rangle = |\Psi(0)\rangle + |\Psi(t_d)\rangle, \quad (8)$$

where

$$|\Psi(0)\rangle = \frac{1}{\sqrt{2}} \sum_j c_j |j\rangle e^{-(\Delta_j/\Delta)^2} e^{-i\omega_j t},$$

$$|\Psi(t_d)\rangle = \frac{1}{\sqrt{2}} \sum_j c_j |j\rangle e^{-(\Delta_j/\Delta)^2} e^{-i\omega_j t} e^{-i(\omega_j - \omega_g)t_d} \quad (9)$$

and the following abbreviations have been used:

$$c_j = \sqrt{\frac{\pi}{\ln 2}} \langle j|\boldsymbol{\mu}\cdot\mathbf{E}_0|g\rangle,$$

$$\Delta = \frac{\sqrt{8\ln 2}}{\tau}, \quad (10)$$

$$\Delta_j = \omega_l - (\omega_j - \omega_g).$$

c_j is proportional to the dipole integral from the ground state $|g\rangle$ to the state $|j\rangle$, Δ represents an energy window of states that can be excited, and Δ_j is the difference between the laser frequency and the frequency associated with the atomic transition from $|g\rangle$ to $|j\rangle$.

Substituting the above expressions for the wave functions into the expression for the modulation amplitude $[M = |\langle\Psi(0)|\Psi(t_d)\rangle|]$ yields

$$M = \left| \sum_j |c_j|^2 e^{-2(\Delta_j/\Delta)^2} e^{i\Delta_j t_d} \right|. \qquad (11)$$

The function can be calculated both semiclassically and quantum mechanically. We have used quantum theory to evaluate it.

C. Evaluation of the autocorrelation function for an atom in parallel electric and magnetic fields

A Gaussian pulse only excites, with any significant probability, a group of quantum states within an energy range of the order $1/\tau$ about the central excitation energy, which is given by the laser frequency ω_l. To evaluate Eq. (11) for a given pulsewidth τ, only the energies and the dipole integrals very near to the frequency of the laser will therefore need to be evaluated.

The Hamiltonian for hydrogen in parallel electric and magnetic fields directed along the z axis is

$$H = -\frac{1}{2}\nabla^2 - \frac{1}{r} + \frac{1}{2}\beta^2\rho^2 + fz, \qquad (12)$$

where β is the magnetic field strength and f is the electric field strength in atomic units. The linear Zeeman term has been omitted because L_z is a conserved quantity for this Hamiltonian and hence m the corresponding quantum number is conserved. For hydrogen, the eigenvalues ω_j and eigenfunctions $|j\rangle$ of this Hamiltonian are evaluated for given fields by expanding the wave function in a basis set

$$\Psi = \sum_{nl} c_{nl} \frac{S_{nl}^\zeta(r)}{r} Y_{lm}(\theta,\phi), \qquad (13)$$

i.e., in products of Sturmian functions and spherical harmonics. The fields have a negligible effect on the ground state $|g\rangle$, which is taken to be the hydrogenic ground state. Once the states $|j\rangle$ have been found the dipole integrals $\langle j|\boldsymbol{\mu}\cdot\mathbf{E}_0|g\rangle$ are readily calculated, hence all of the quantities entering Eq. (11) are known.

For rubidium, the method developed by O'Mahony and Taylor [15] for nonhydrogenic atoms in fields is used. The Hamiltonian in Eq. (12) is only valid at a radius, $r = b$, outside of the atomic core and hence the expansion in Eq. (13) is now employed over a semi-infinite region $r = b$ to ∞. The eigenvalues and eigenfunctions of H plus a surface term at $r = b$ are used to construct an R matrix or log-derivative matrix at $r = b$. This R matrix is matched to phase-shifted Coulomb functions at $r = b$ and an energy search is conducted to find eigenenergies ω_j such that the total wave function and its derivative are continuous over the boundary. The corresponding eigenvectors $|j\rangle$ can then be constructed. Note that it is at this stage that the fingerprint of the nonhydrogenic core is introduced via the quantum defects used to calculate the phase-shifted Coulomb functions.
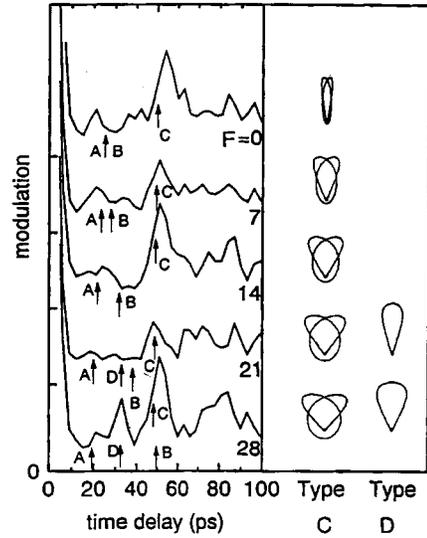

FIG. 1. Experimental results of Marmet *et al.* [4] showing the magnitude of the autocorrelation function M as a function of the time delay between pulses for a rubidium atom in parallel electric and magnetic fields. The magnetic field is constant at 1.5 T and a static electric field F is varied as indicated. The periods of the classical orbits A, B, C, and D are indicated. A and B are orbits along the $+z$ and $-z$ directions, respectively, and the orbits C and D are shown on the right-hand side of the diagram.

III. RESULTS

The experimental amplitude M as a function of the time delay between the two pulses is shown in Fig. 1 [4]. The rubidium atom is excited from its ground state by the pulse sequence using linearly polarized light and in the presence of a fixed magnetic field of $B = 1.5$ T and a static electric field ranging from $F = 0$ to 28 V/cm. (Note that $\beta = B/B_c$ and $f = F/F_c$, where $B_c = 4.7\times10^5$ T and $F_c = 5.14\times10^9$ V/cm.) The frequency of the laser is such that final Rydberg states centered around the binding energy -36.1 cm^{-1} are excited by the pulses. For $F = 0$, this energy range corresponds classically to regular motion and quantum mechanically to the region where the external fields are but a perturbation. However, as F increases one goes over to the strong field mixing region where the quantum spectrum becomes irregular and the classical mechanics becomes chaotic. Figure 1 shows the appearance of broad peaks at certain time delays. These times correspond to the shortest periodic orbits in the system. For specific values of the static fields, the periods of these orbits can be determined from the classical equations of motion. Four of these periods are indicated on the diagram by the letters A to D. The periodic orbit designated by A is the orbit parallel to the z axis while B is the orbit antiparallel to the z axis. The orbit C is shown in Fig. 1 as well as the orbit D, which results from a bifurcation of the orbit B at about 20 V/cm.

To examine these results theoretically, M must be determined as a function of the time delay for hydrogen in parallel electric and magnetic fields. Equation (11) is evaluated for hydrogen, excited from its ground state in a fixed magnetic field $B = 1.5$ T and for static electric fields ranging from 0 to 28 V/cm (see Fig. 2). Using linearly polarized light the pulse excites the atom from its ground state to final Rydberg states
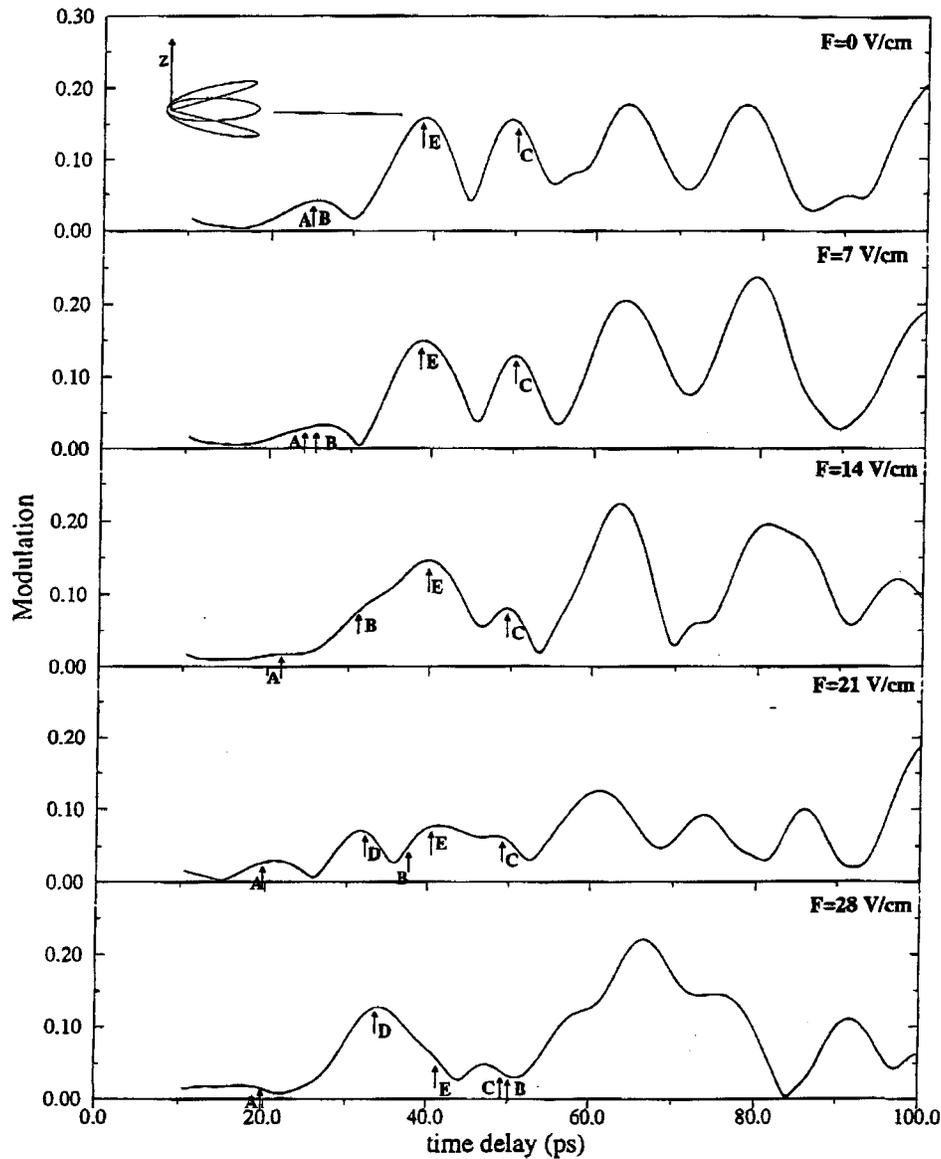
Hydrogen

FIG. 2. Theoretical results showing the magnitude of the autocorrelation function M as a function of the time delay between pulses for a hydrogen atom excited from the ground state in parallel electric and magnetic fields. The magnetic field strength is 1.5 T in every panel and the electric field strength F varies from 0 to 28 V/cm. The letters mark the periods of the classical orbits as in Fig. 1 and the orbit E is indicated in the inset.

centered around a total energy of -36.1 cm^{-1}. The pulse-width τ is taken to be 4 ps. The polarization of the laser light and the fact that L_z is conserved means that only $m=0$ final states need to be considered in the basis set expansion in Eq. (13). The condition that the Gaussian energy window $\exp[-2(\Delta_j/\Delta)^2]$ decays off to a negligible value, requires that all Rydberg states within about 7 cm^{-1} of the above total energy need to be calculated. The relevant energy levels and dipole integrals are obtained by the basis set methods described above and hence M can be evaluated. The sum in Eq. (13) is taken to be large enough to achieve converged states in this energy range, and the common exponent ζ is

varied to check convergence of the energy levels. These energy levels are used to calculate the corresponding amplitude from Eq. (11), for time delays from 0 to 100 ps. Broad peaks are observed at certain time delays. As in Fig. 1 the periods of the orbits A, B, C, and D are shown as a function of the electric field strength F. The bifurcation of the orbit B into D is confirmed by our results, however, an additional peak, labeled E, is also seen at about 36 ps. This peak, which is not present in the experiment, can be identified with the classical orbit shown in the inset.

To simulate the experiment and to examine core effects M was also calculated for excitations from the rubidium

3728 MOSER, MOTA-FURTADO, O'MAHONY, AND dos SANTOS
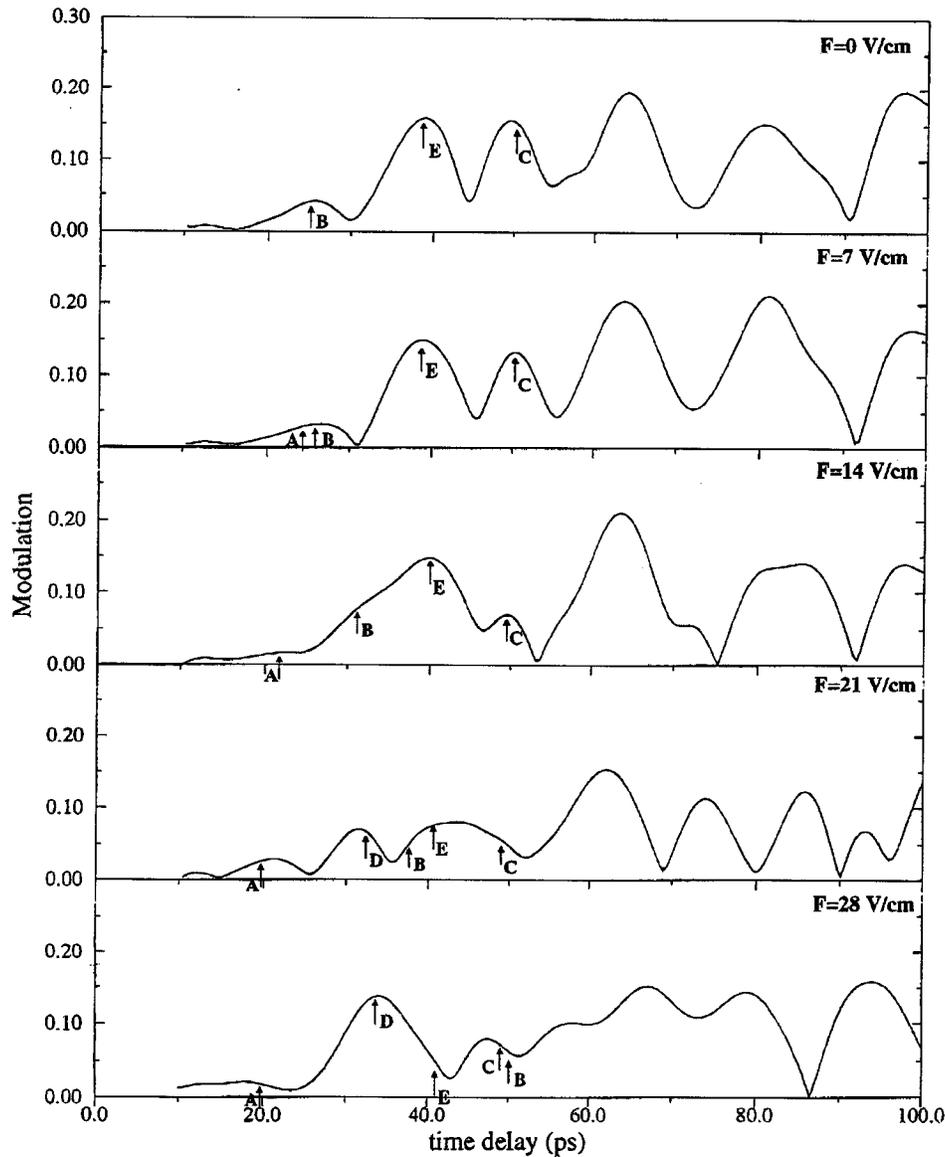
Rubidium

FIG. 3. Theoretical modulation spectra for rubidium in parallel electric and magnetic fields with the same parameters as in Fig. 2.

ground state. The energy levels of rubidium in external electric and magnetic fields are obtained by employing the R-matrix approach [16]. In the experiment the rubidium ground-state hyperfine levels are thermally populated. However, the small energy differences between these states in the presence of the external fields only induce effects in Eq. (11) for time delays of the order of 100 ps and can therefore be neglected in our analysis [5]. Energy levels and oscillator strengths for rubidium are used in the evaluation of Eq. (11) and the results are presented in Fig. 3. A qualitatively similar picture to hydrogen is obtained for short time delays. The main difference between hydrogen and rubidium, and hence the effect of the core, is for long time delays $t_d > 60$ ps as has been seen previously in Fourier transform of the frequency domain spectra [16]. These differences can be viewed as

being due to scattering of the wave packet from the core for longer time delays.

The discrepancies between theory and experiment cannot be explained by the rubidium core as this has been fully included via the R-matrix method. There are several possible explanations for the differences between the experimental and theoretical time delay spectra. In the experimental paper [4] it is mentioned that a poor signal-to-noise ratio is obtained when the laser field is parallel to the magnetic field as in the case studied here. Also any errors in the pulse shape may alter the spectrum obtained because of the different distribution of excited Rydberg levels. It is clear that while there may be several experimental problems to overcome this phase sensitive technique provides a very promising method to observe directly in the time domain the frequen-

cies of the shortest periodic orbits for classically chaotic systems in general.

In summary the theoretical results reproduce qualitatively the main features of the experiment. However, detailed quantitative agreement is lacking. The theory also gives an extra peak for hydrogen and rubidium associated with the orbit of period 36 ps, shown in the inset of Fig. 2.

[1] J. Main, G. Weibusch, A. Holle, and K.H. Welge, Phys. Rev. Lett. **57**, 2789 (1986).

[2] A. Holle, J. Main, G. Weibusch, H. Rottke, and K.H. Welge, Phys. Rev. Lett. **61**, 161 (1988).

[3] J.A. Yeazell, G. Raithel, L. Marmet, H. Held, and H. Walther, Phys. Rev. Lett. **70**, 2884 (1993), and references therein.

[4] L. Marmet, H. Held, G. Raithel, J.A. Yeazell, and H. Walther, Phys. Rev. Lett. **72**, 3779 (1994).

[5] G. Raithel, H. Held, L. Marmet, and H. Walther, J. Phys. B **27**, 2849 (1994).

[6] J. Wals, H.H. Fielding, J.F. Christian, L.C. Snoek, W.J. van der Zande, and H.B. van Linden van den Heuvell, Phys. Rev. Lett. **72**, 3783 (1994).

[7] B. Broers, J.F. Christian, and H.B. van Linden van den Heuvell, Phys. Rev. A **49**, 2498 (1994).

[8] H.H. Fielding, J. Wals, W.J. van der Zande, and H.B. van Linden van den Heuvell, Phys. Rev. A **51**, 611 (1995).

[9] G.M. Lankhuijzen and L.D. Noordam, Phys. Rev. A **52**, 2016 (1995).

[10] J. Parker and C.R. Stroud, Phys. Rev. Lett. **56**, 70 (1986).

[11] L.D. Noordam, D.I. Duncan, and T.F. Gallagher, Phys. Rev. A **45**, 4734 (1992).

[12] J.F. Christian, B. Broers, J.H. Hoogenraad, W.J. Van der Zande, and L.D. Noordam, Opt. Commun. **103**, 79 (1993).

[13] S. Tomsovic and E.J. Heller, Phys. Rev. E **47**, 282 (1993), and references therein.

[14] G. Alber and P. Zoller, Phys. Rep. **199**, 231 (1991).

[15] P.F. O'Mahony and K.T. Taylor, Phys. Rev. Lett. **57**, 2931 (1986).

[16] P.F. O'Mahony, Phys. Rev. Lett. **63**, 2653 (1989).