Observation of Wave Packet Motion along Quasi-Landau Orbits

J. A. Yeazell, G. Raithel, L. Marmet, H. Held, and H. Walther
Max-Planck-Institut für Quantenoptik and Sektion Physik der Universität München, D-8046 Garching, Germany
(Received 25 January 1993)

The evolution of wave packets is investigated in crossed magnetic and electric fields. We report a direct observation of their propagation along classical trajectories in the time domain. Rydberg states of rubidium are excited with a short laser pulse. We monitor the evolution of the wave packet using a delayed, ionizing probe pulse. When the wave packet approaches the nucleus, the ionization signal is enhanced. The first return to the nucleus is observed and is in agreement with semiclassical predictions. Further evolution of the wave packet is also observed.

PACS numbers: 32.60.+i, 32.30.Jc, 35.10.Di

The most fundamental system of atomic physics is the hydrogen atom. It is also the most thoroughly studied and best understood. However, when it is brought into a strong magnetic field, the properties of this new atom-field system become quite different. For hydrogenic Rydberg atoms, it is possible to access experimentally the interesting regime, where the Coulomb interaction and the diamagnetic interaction are comparable. Spectral studies of this system find significant changes in the structure in this regime near the ionization threshold. In particular, Garton and Tomkins [1] discovered a sinusoidal modulation of an extremely dense spectrum. These sinusoidal oscillations are commonly known as quasi-Landau (QL) resonances. It was first recognized by Edmonds [2] and Starace [3] that the QL resonances are associated with classical closed trajectories of the system. In this paper, we focus on these classical characteristics and report a direct observation of the classical dynamics of this atom.

The central subject of classical mechanics is defining the position of a particle as a function of time. Of particular interest to our present work is the now established ability to produce nonstationary superpositions of Rydberg states whose wave packets exhibit the proper classical dynamics. These classical-like atomic states are similar to the coherent states of the harmonic oscillator.

Experiments have been carried out that produce a variety of classical-like states, e.g., radially localized [4–7], angularly localized [8], and Stark state wave packets [9]. The simplest to describe is the radial wave packet. In this case, a short optical pulse interacts with the ground state of a single-electron atom producing a superposition of Rydberg states with a distribution of the principal quantum number. However, the superposition state contains only a single value for the angular momentum quantum number and for the Zeeman quantum number. The resulting excited state wave packet is spatially localized solely in the radial coordinate and it oscillates between the nucleus and the classical outer turning point. Subsequent interactions with this wave packet are modified by this temporal evolution. It is only when the wave packet is near the nucleus that further optical interactions, such as photoionization of the classical-like state, can take place. The excitation of the other types of classical-like states requires the application of appropriate additional fields. The production of wave packets in strong, external fields has been considered theoretically [10–13]. Here, we study such classical-like states experimentally.

The QL spectrum mentioned is a member of a larger class of phenomena. Other types of QL resonances have been observed in magnetic fields as well as in crossed electric and magnetic fields [14–17]. These other types of QL resonances also are related to classical recurring trajectories. Examples of some typical QL spectra are shown in Fig. 1. Their envelopes also display periodic structures. The theoretical treatment of this problem is extensive ([18–21] and references herein). These semi-

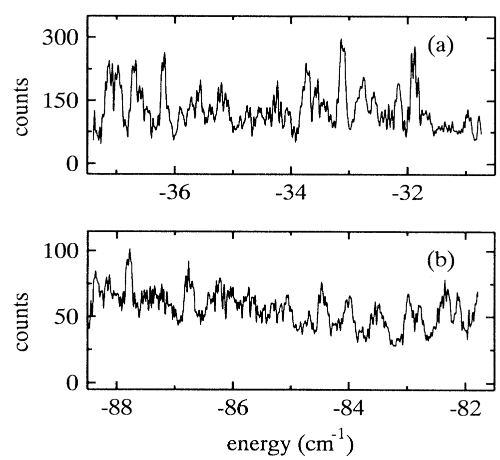

FIG. 1. Quasi-Landau spectra in crossed fields. These spectra are energy averaged to highlight the slow modulations of the envelope. In (a) the magnetic field strength was 2.14 T and the electric field was 2500 V/m. At the average radial value of the dominant orbit, $r \approx 2000a_0$, the diamagnetic energy is 4×10^{-5} a.u., 4 times larger than the Stark term, and 12 times smaller than the Coulomb term. In (b) the magnetic field strength is 0.84 T and the electric field strength was 16 000 V/m. At $r \approx 2000a_0$, the diamagnetic energy is 6×10^{-6} a.u., 10 times smaller than the Stark term, and 80 times smaller than the Coulomb term.

classical treatments of the problem accurately predict the structure of the envelope of the observed energy-averaged spectra. Currently, they do not make predictions about the detailed structure beneath the envelope.

The studies of the quasi-Landau regime have been confined to the frequency domain. Here, we extend these studies to the time domain by directly observing the classical dynamics of wave packets excited in such a system. Because of the high symmetry of the Coulomb potential, the wave packets obtained in zero external field correspond to a one- or two-parameter family of classical orbits, e.g., a radial wave packet. A magnetic field alone does not completely remove the symmetry; it removes only the symmetry associated with the azimuthal angle. However, in the case of crossed electric and magnetic fields, no continuous symmetry is left. In such crossed fields it is possible to excite a wave packet that is localized in all spatial dimensions. It is precisely this close classical analog that is studied in the following experiment. The results are interpreted classically and compared with a semiclassical theory.

The basic concept of the experiment is straightforward. A short ultraviolet pulse excites a superposition of states located beneath the envelope of the QL spectra; the bandwidth of the pulse spans the spectra displayed in Fig. 1. The resulting wave packet has the periodicities of the classical trajectories of this system. The spatial localization of the wave packet increases as more modulations of the envelope are overlapped. However, if the excitation pulse is too short, then the frequency of the modulations and their amplitude can change over the bandwidth of the pulse producing a wave packet that disperses rapidly. Ideally the pulsewidth should be approximately an order of magnitude smaller than the orbital period. The wave packet's spatial localization and temporal evolution modify its interaction with a subsequently applied probe laser. As in the case of the radial wave packet, the ionization process driven by the probe laser is significant only when the wave packet approaches the nucleus. By adjusting the delay time between the pump and probe laser pulses, the temporal evolution of the wave packet can be mapped out.

It is important to note a significant difference between the wave packet excited in this crossed field system and the field-free radial wave packet. The radial wave packet could be thought of as a superposition of identical classical orbits with arbitrary direction of the Runge-Lenz vector. In contrast, the quasi-Landau wave packet can consist of strikingly different types of classical orbits, each with its own distinct orbital period. Such a wave packet would evolve as a collection of subwave packets along all these trajectories simultaneously. However, for certain parameters of the quasi-Landau system, a single orbit type does dominate. Experimentally this type of quasi-Landau wave packet produces the clearest signal.

The experimental setup, which is similar to that described in [17], is sketched in Fig. 2. A beam of rubidium atoms effusing from an atomic beam oven (nozzle diameter ≈ 0.3 mm) enters a field region providing homogeneous crossed electric and magnetic fields, the latter being parallel to the atomic beam direction. The superconducting magnet produces a maximum magnetic field strength of 6 T at the excitation point.

The necessary short laser pulses are obtained from a single 594 nm pulse formed in a synchronously pumped R6G dye laser. The pump laser is a cw-mode-locked Ar^+ laser with an 80 MHz repetition rate. The dye laser is cavity dumped by an intracavity acousto-optic modulator which selectively extracts every 25th yellow pulse (8 ps FWHM, 10 nJ). The resulting repetition rate of the dye laser is approximately 3 MHz. An ultraviolet pulse of 6 ps duration is obtained by second-harmonic generation in a $LiIO_3$ crystal with 10% conversion efficiency. This UV pulse is separated from the yellow pulse by a dichroic mirror. The yellow pulse traverses an adjustable delay line before being recombined with the UV pulse. Both pulses are focused into the interaction volume by a single 30 cm lens. However, an additional pair of lenses in the delay arm adjusts the divergence of the yellow beam so that the focal spots (≈ 50 μm) of the UV pump and yellow-probe pulses overlap. In the following experiments, both optical fields were polarized parallel to the electric field.

The combination of the magnetic field and the electric field electrodes make up a modified Penning trap which temporarily stores the electrons produced from the photoionization. Eventually all the electrons approach an auxiliary electrode which extracts them from the trap. The magnetic stray field images the extraction point onto the surface of a microchannel plate detector (MCP). This trapping of the photoelectrons greatly increases our col-

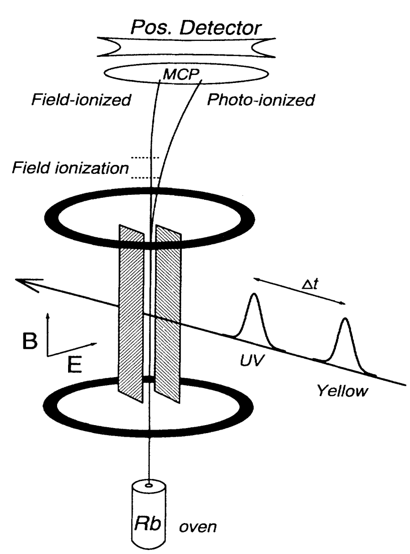

FIG. 2. Sketch of the experimental setup.

lection efficiency.

The Rydberg atoms which are not photoionized by the yellow pulse leave the excitation field region, and are field ionized in a separate, approximately vertical electric field. The corresponding field ionization point also is imaged by the magnetic stray field onto the MCP detector. Thus, on the MCP surface there are two well-resolved spots where electrons impact: one spot corresponding to the field-ionized Rydberg atoms, and the other spot corresponding to atoms which have been photoionized already in the excitation field region. A Hamamatsu position sensitive detector allows the signals from these two spots to be recorded separately.

The time delay between the UV pulse and the yellow pulse is stepwise scanned, and for each step the counts in the two spots are integrated. The average count in a 1 ps time bin is 1500. The signal-to-noise ratio is improved by recording a background signal where the yellow light is blocked by a shutter. This background signal is averaged and used to remove long-term variations of the signal. The signal-to-noise ratio may be further improved by taking advantage of correlations between the field-ionized signal and the photoionized signal. We are only interested in the process which photoionizes Rydberg atoms. Such a process leads to an increase in the photoelectrons at the expense of the Rydberg atom population. The subtraction of the field-ionized signal from the photoionized signal emphasizes this anticorrelation. At the same time, this procedure reduces short-term fluctuations, since the ions and the stable atoms are counted simultaneously.

A simple semiclassical theory can be formulated to model our experiment. We are interested in the two-photon process where a UV photon excites the Rydberg state and after a suitable delay a yellow photon ionizes it. The theoretical study of Rydberg atom wave packets in strong fields [10–13] provides a semiclassical approximation to the two-photon transition matrix element needed to determine the ionization probability. A semiclassical Green's function is utilized for this process which is valid outside a sphere of about $50a_0$. An ensemble of electron trajectories provides the propagation away from and back to this surface. The sensitivity of a particular trajectory to its starting angles determines the amplitude of the associated recurring wave. As the sensitivity increases the amplitude decreases. Within this sphere the wave function is described by a sum of Coulomb waves, the amplitudes of which are found by matching with the returning semiclassical waves at the surface. The total two-photon matrix element is written as a sum of the contributions from different recurring trajectories. The photoionization probability as a function of the delay time gets large if the delay time coincides with the time of traversal of a strongly modulating orbit. Last, the laser polarization plays a significant role in determining the type of trajectories that may be excited. Here, a polarization parallel to the static electric field is used to enhance the excitation of stable orbits, i.e., those which are most insensitive to the initial angles.

The present form of the semiclassical model correctly identifies the dominant orbits which produce the first recurrence. However, we find that these orbits suffer a strong collision with the nucleus at the end of one orbit. Here our experiment differs from one using hydrogen. Beyond the first recurrence, the scattering produced by the rubidium core makes further predictions of the evolution nontrivial. Our current model also does not include dispersion of the wave packet (the action is linearly approximated in the energy). Finally, interference effects between different types of orbits which have similar times of traversal are not taken into account. The goal of this simple model is the accurate description of the first recurrence.

In Fig. 3(a), the delay time signal is displayed in a regime where the magnetic field is dominant. The wave packet is excited for the same parameters as the spec-

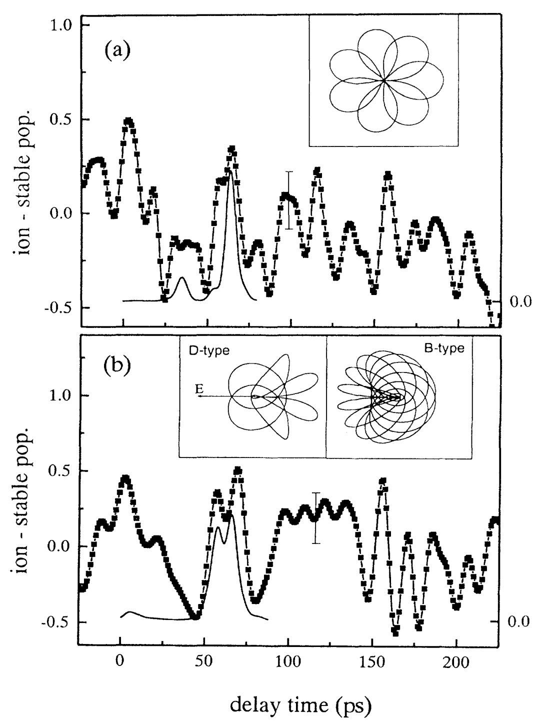

FIG. 3. The difference between the photoionized signal and the field-ionized signal vs delay time. Also shown is the semiclassical prediction of the transition probability for the two-photon ionization process that consists of an ultraviolet pump pulse exciting the Rydberg states and delayed, yellow pulse ionizing these states. The trajectories of the dominant orbits are shown in the insets. The parameters are the same as in Fig. 1: (a) the strong magnetic field case; (b) the strong electric field case. In (b), two orbits dominate the behavior. The D type starts in the direction of the electric field ionization saddle point and the B type starts in the opposite direction.

trum shown in Fig. 1(a). Also shown on the plot are the predictions of the ionization probability derived from the semiclassical analysis. The peak at zero delay time is due to the simultaneous presence of both pump and probe pulses. Another large peak occurs when the wave packet has completed an orbital cycle and is again near the nucleus (at 62 ps). This first return to the nucleus is in excellent agreement with the predicted classical orbital period of the dominant orbit for this range of parameters [shown in the inset of Fig. 3(a)]. That is, the semiclassical transition probability is peaked at the same time as the experimental signal. Further oscillations of the experimental signal are evident after the first return. The general broadening of the signal and the appearance of higher frequency oscillations are characteristics consistent with dispersion and scattering of the wave packet. A Fourier transform of the spectrum displayed in Fig. 1(a) finds a major peak at the orbital periods of 62 ps and two subsidiary peaks at 97 ps and 18 ps. These results are in agreement with this time domain study. The error is estimated by the variation of the signal upon repetition of the experiment.

In Fig. 3(b) the delay time signal is now shown in a regime where the electric field is dominant. The experimental parameters correspond to the spectrum shown in Fig. 1(b). Here, again the first return of the wave packet is seen clearly as a double-peaked structure at 58 ps and 67 ps. This double-peaked structure is also evident in the semiclassical transition probability. Classically, these two peaks are attributed to similar type orbits with slightly different orbital periods. The analogous wave packet then consists primarily of two subwave packets following these orbits. The Fourier transform of the energy spectrum is consistent with this delay-time signal. It displays two equally strong peaks at 58 and 67 ps. Following the first recurrence we again see a rapid broadening of the oscillations and a growth of higher frequency oscillations.

The observation of the evolution of the wave packet establishes a connection between the time domain and the previous frequency domain studies of quasi-Landau systems. The trajectories corresponding to the observed times of traversal of the wave packets coincide with the orbits which lead to the dominating quasi-Landau resonances. Similarly, the behavior in the time domain can be predicted, at least up to the first recurrence, by semiclassical arguments. Further evolution is complicated by the scattering of the wave packet off the core potential, and by dispersion and quantum interference effects. Neither of these effects is included in our model. The development of a more complete theory lies outside the scope of this paper. It should be noted that many of the experimental peaks following the first recurrence occur at times which are sums of the dominant orbital periods. This is particularly clear in the high magnetic field regime. Scattering is expected to be strong in this case and this suggests that these sum periods are due to subsequent propagation along two (or more) primitive recurring orbits, whereby successive orbits are connected via intermediate scattering at the core potential.

The wave packet created in this system is somewhat unique. The lack of continuous symmetry in the system requires that the wave packet be localized in three dimensions. This would be the first experimental excitation of such a particlelike atomic wave packet. Unfortunately, a direct measure of this spatial localization is difficult to obtain.

One of us (L.M.) would like to acknowledge the support of the Alexander von Humboldt Foundation.

[1] W.R.S. Garton and F.S. Tomkins, Astrophys. J. **158**, 839 (1969).
[2] A.R. Edmonds, J. Phys. (Paris), Colloq. **31**, C4-71 (1970).
[3] A.F. Starace, J. Phys. B **6**, 585 (1973).
[4] A. ten Wolde, L.D. Noordam, H.G. Muller, A. Lagendijk, and H.B. van Linden van den Heuvell, Phys. Rev. Lett. **61**, 2099 (1988).
[5] J.A. Yeazell, M. Mallalieu, J. Parker, and C.R. Stroud, Jr., Phys. Rev. A **40**, 5040 (1989).
[6] J.A. Yeazell, M. Mallalieu, and C.R. Stroud, Jr., Phys. Rev. Lett. **67**, 2007 (1990).
[7] J.A. Yeazell and C.R. Stroud, Jr., Phys. Rev. A **43**, 5153 (1991).
[8] J.A. Yeazell and C.R. Stroud, Jr., Phys. Rev. Lett. **60**, 1494 (1987).
[9] A. ten Wolde, L.D. Noordam, A. Lagendijk, and H.B. van Linden van den Heuvell, Phys. Rev. A **40**, 485 (1989).
[10] G. Alber, Phys. Rev. A **40**, 1321 (1989).
[11] G. Alber, Z. Phys. D **14**, 307 (1989).
[12] G. Alber, Comments At. Mol. Phys. **26**, 47 (1991).
[13] G. Alber and P. Zoller, Phys. Rep. **199**, 231 (1991).
[14] A. Holle, G. Wiebusch, J. Main, B. Hager, H. Rottke, and K.H. Welge, Phys. Rev. Lett. **56**, 2594 (1986).
[15] J. Main, G. Wiebusch, A. Holle, and K.H. Welge, Phys. Rev. Lett. **57**, 2789 (1986).
[16] G. Wiebusch, J. Main, K. Krüger, H. Rottke, A. Holle, and K.H. Welge, Phys. Rev. Lett. **62**, 2821 (1989).
[17] G. Raithel, M. Fauth, H. Walter, Phys. Rev. A **44**, 1898 (1991).
[18] M.C. Gutzwiller, Physica (Amsterdam) **5D**, 183 (1982).
[19] M.L. Du and J.B. Delos, Phys. Rev. A **38**, 1896 (1988).
[20] M.L. Du and J.B. Delos, Phys. Rev. A **38**, 1913 (1988).
[21] H. Friedrich and D. Wintgen, Phys. Rep. **183**, 37 (1989).