Stueckelberg Oscillations in the Multiphoton Excitation of Helium Rydberg Atoms: Observation with a Pulse of Coherent Field and Suppression by Additive Noise

S. Yoakum, L. Sirko,[a] and P. M. Koch

Physics Department, State University of New York at Stony Brook, Stony Brook, New York 11794-3800

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We observe and reproduce theoretically Stueckelberg oscillations in the microwave excitation of helium ($n=28$) ^3S Rydberg atoms. They are produced by the coherent evolution along two isolated Floquet potential curves as the microwave electric field amplitude varies along a half-sine-wave pulse shape. The measured visibility and separation of the oscillations depends sensitively on the Floquet structure, on the atomic beam velocity, and particularly on additive noise, which smears and broadens the pattern.

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In the early 1930s Landau [1], Zener [2], and Stueckelberg [3] laid the theoretical foundations for curve-crossing problems in the context of slow atomic collisions. Their approach exploited the existence of two disparate time scales (identified for molecules by Born and Oppenheimer [4])—one associated with the fast electronic motion, and the other with the slow variation of the internuclear coordinate **R** [5]. Calculating the electronic structure at many fixed values of **R** yields a potential curve for each of the molecular states. During a slow collision, the states of the molecular complex evolve along these curves as **R** varies. In this picture the evolution is largely adiabatic, and nonadiabatic behavior is restricted to narrow regions in **R** where two or more potential curves anticross.

Depending on the geometry of the anticrossing curves and the relative velocity $d\mathbf{R}/dt$ of the approaching collision partners, an anticrossing may be traversed adiabatically, diabatically, or in a coherent mixture of the two [6]. In the latter case the probability amplitudes evolving along two potential curves interfere when the same anticrossing is traversed again as the collision partners separate. As a function of the collision energy, the variation of the difference in dynamical phase evolution [7] between the anticrossings produces oscillations in the final-state populations. Because Stueckelberg was the first to include the effects of this phase coherence in the collision theory, they are called Stueckelberg oscillations.

The same formalism may be applied to the study of Rydberg atoms in oscillatory electric fields by utilizing the Floquet theorem and Fourier methods [8]. In this framework the atom plus microwave field system evolves along quasienergy levels, or equivalently Floquet potential curves, as the microwave field amplitude varies slowly along its pulse shape [9]. Earlier theoretical work predicted that Stueckelberg oscillations should be observable as the peak field strength is slowly scanned beyond an anticrossing [9].

In this Letter we report the first observation of Stueckelberg oscillations in the microwave excitation of helium [10]. To observe them, several conditions must necessarily be met simultaneously: An anticrossing between otherwise isolated quasienergy states must be found such that the anticrossing geometry, the field pulse shape, and the atomic beam velocity together lead to a diabatic transition probability through the crossing of order $\frac{1}{2}$. These same factors must also be such that as the peak field amplitude is scanned, the growth rate of the phase difference between the two traversals of the anticrossing is slow enough to make the interference oscillations resolvable. Finally, a high level of coherence of the microwave field must be maintained, for as our results show, additive noise can completely destroy the oscillations.

Figure 1 presents a Floquet map showing the quasienergy structure calculated for the $n=28$, $M_L=0$, spin triplet manifold at many fixed amplitudes of a 29-GHz microwave field. How the map is constructed and the general features of helium quasienergy structure have been discussed previously [9]. For this map, the basis set included the nearby $n=29$, $L=0\rightarrow 4$, and $n=27$, $L=26$ triplet states in addition to the $n=28$ manifold. We found empirically that thirty photon blocks provided a satisfactory level of convergence. Throughout this Letter, quasienergy states in the microwave field will be labeled by the quantum numbers appropriate to the zero-field states to which they are diabatically connected. The notation is (n,L,k), with k representing the relative photon number. Note that the Floquet structure is periodic in energy and that since the total angular momentum of an isolated atom plus field system must be conserved, the period in the map is twice the photon energy ($T=2\times 0.96733$ cm^{-1}). In other words, a Floquet map needs only to contain quasienergy states of the same parity $(-1)^{L+k}$ [here $+1$ because the initial state is (28,0,0), as will be described below]. Also note that the curvature of the Floquet levels with field amplitude (dynamic polarizability) is largest in those (nonhydrogenic) states which would exhibit large second-order Stark shifts in a dc electric field but is very small for hydrogenic states (small quantum defects). Finally, it must be emphasized that perfect coherence of the microwave field is inherent to a Floquet map.

Figure 1 shows an isolated anticrossing at 34.5 V/cm between the (28,0,4) and (28,1,1) even-parity states [11].

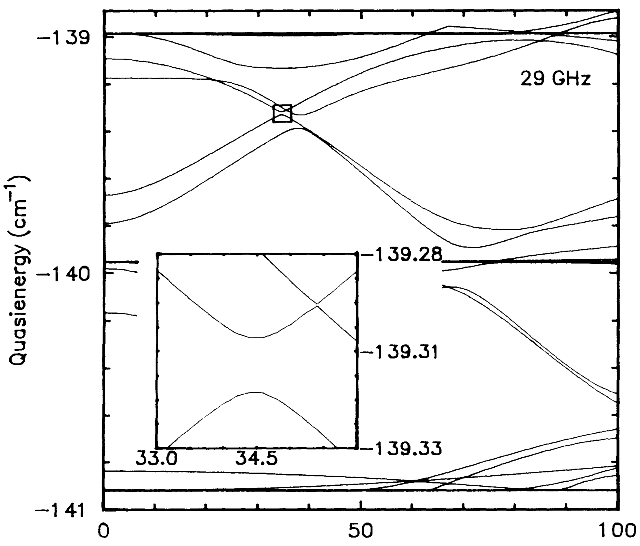

FIG. 1. Map of even-parity quasienergy levels relevant for the $n=28$, $M_L=0$ triplet helium manifold at 29 GHz. Inset: The anticrossing at 34.5 V/cm between (28,0,4) (upper) and (28,1,1) (lower) levels.

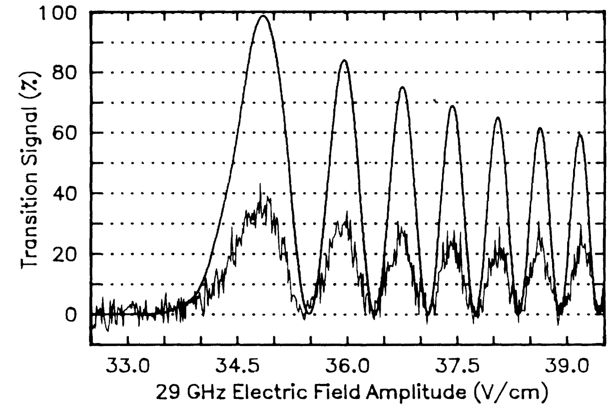

FIG. 2. Probability for microwave-induced transition from the initial $28\,^3S$ state to $28\,^3P$ vs the peak field amplitude F_0. Jagged curve: Experimental data for 16-keV beam. Smooth curve: Integration of the Schrödinger equation on a two-state model using a half-sine-wave pulse shape. See the text for a discussion of the vertical scales.

We have modeled the microwave-pulse-shape-driven dynamics on this structure by an integration of the Schrödinger equation. The integration began with unit population on the $28\,^3S$ state and used a half-sine-wave pulse shape for the 29-GHz microwave field amplitude. After the interaction, the population in the $28\,^3P$ state was evaluated. For each of many peak field amplitudes, the integration was repeated, producing the interference pattern of Stueckelberg oscillations shown as the smooth, solid curve in Fig. 2. In numerical results, when the field pulse lies just beyond 34.5 V/cm, nearly 100% of the population is transferred to the P state. Moreover, the separation between the first few oscillations is about 1 V/cm. Inspired by this result, which was the best obtained from many calculations carried out for a number of different n values and frequencies, we performed the experiment described below.

As described elsewhere [9], we used a fast-beam apparatus and two-step $^{12}C^{16}O_2$-laser excitation to produce a monoenergetic ($\Delta v/v \simeq 0.003$) beam of helium atoms in the $28\,^3S$ Rydberg state. The microwave interaction took place inside a section of Ka-band (26–40 GHz) waveguide that had a 0.53-mm-diam hole machined in each short sidewall for beam entrance and exit [12]. After the microwave interaction we measured the population of atoms that had been transferred to harder-to-ionize states (compared to $28\,^3S$) using standard selective (static) field-ionization techniques.

To achieve the half-sine-wave pulse shape for a 29-GHz microwave field, we used a traveling-wave system. The 14.5-GHz output of a Gigatronics model 900 synthesized microwave source was amplified with a Miteq model AFD4-080180-20P preamp, frequency doubled with a Honeywell model A2000N doubler, and amplified again with a 1-W Litton model 1077H12F00 26–40-GHz traveling-wave-tube amplifier (TWTA) to deliver microwave power to the interaction region. The waveguide, operating in the TE_{10} mode, crossed the atomic beam at right angles and was terminated with a well-matched (measured voltage standing-wave ratio of ≤ 1.0001) absorbing ferrite load inserted directly into the copper waveguide section inside the vacuum. To prevent outgassing of the load heated by the microwaves, we used powers below 300 mW.

A fraction fP_{inc} of incident microwave power was monitored outside the vacuum system with a Marconi model 690 power meter and model 6913 power head mounted on a directional coupler. With the apparatus open to atmosphere, the power in the interaction region was measured using another power meter head mounted in place of the load to determine, along with calculations, a calibration factor $CF=8.71$, which relates fP_{inc} to the 29-GHz field amplitude F_0 in the interaction region: $F_0[\mathrm{V/cm}] = CF(fP_{inc}[\mathrm{mW}])^{1/2}$. We estimated the uncertainty in F_0 to be 5%.

Downstream of the waveguide, a static field of 652 V/cm was applied between a pair of 10-cm-long parallel plates (analysis plates). Empirically we found that this field completely ionized Rydberg atoms in the initial $28\,^3S$ state. However, the probability of ionizing $28\,^3P$ atoms in this field was measured to be only 65% ($\pm 5\%$). The surviving 35% could be ionized further downstream by a much higher field in a longitudinal-field ionizer, with the resulting ions detected with a Johnston MM1 particle multiplier [13]. Thus, if the microwave interaction left all atoms in the initial $28\,^3S$ state, the signal would be zero. On the other hand, should the microwaves transfer

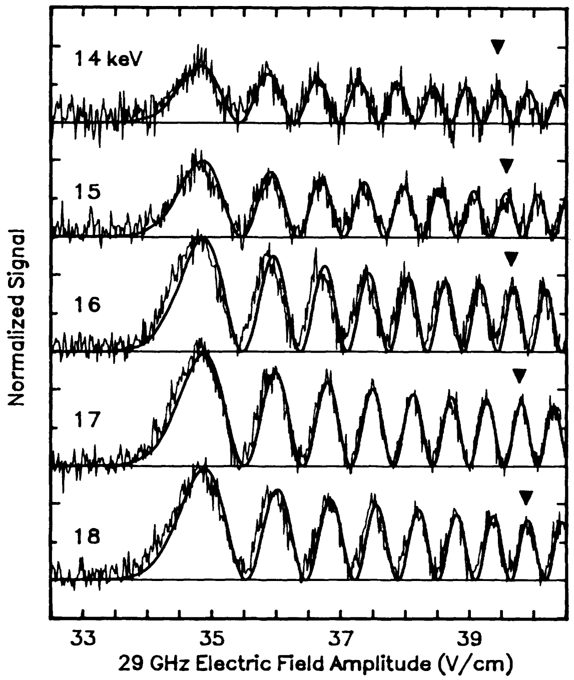

FIG. 3. Experimental data at 29 GHz for five different atomic beam energies in keV. Smooth curves: Numerical integration of the Schrödinger equation for each case. For ease of comparison, each has been separately normalized (vertically) to the data. Solid horizontal lines indicate 0% level for each curve.

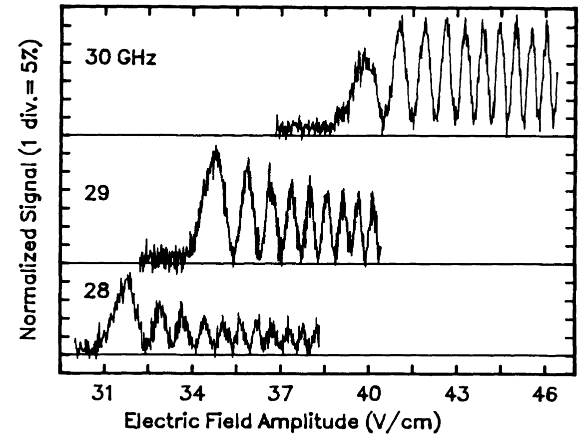

FIG. 4. Experimental data at 16 keV for $28\,^3S$-$28\,^3P$ transitions in a 29-GHz field (with fitted *CF*) and 28- and 30-GHz fields (with empirical *CF*s). Solid lines indicate 0% level.

100% of the initial $28\,^3S$ atoms to the $28\,^3P$ state, the signal would be about 35% of the initial population.

The jagged curve in Fig. 2 shows data collected in a multiscalar as described above, with a 16-keV atomic beam. For normalization purposes, the microwave power was turned off during the first thirty channels of each scan (not shown), and the analysis-plate voltage was also turned off for the first fifteen channels. Thus, the signal measured in the first fifteen channels represented 100% of the initial $28\,^3S$ atoms, while the next fifteen channels represented 0%. The measured pattern registers very well with the model results when a $CF=8.335$ is used. This value differs from the empirical value of 8.71 by 4%, which is within the estimated 5% calibration uncertainty. The vertical heights of the measured oscillations are 35% of the calculated ones, as expected from the relative $28\,^3S$-$28\,^3P$ detection sensitivities.

Varying the beam velocity changed the duration but not the shape of the microwave field pulse seen by the atoms. For a given pulse amplitude F_0, as the beam energy was increased, less phase difference was accrued between the anticrossings. This would cause the interference pattern to be stretched as a function of F_0. The experimental data shown in Fig. 3 bears this out. Note that there was no observable shift in the position of the first peak. This is to be expected because the Floquet structure was not altered. Varying the interaction time by a small amount only caused the interference pattern to be modified. For example, the arrowhead shows the shifting position of the eighth peak.

In contrast, because the Floquet structure is altered when the microwave frequency is changed, we expect the entire pattern to be affected as the frequency is varied. The data in Fig. 4 show $28\,^3S$-$28\,^3P$ Stueckelberg oscillation data collected for three different frequencies with a 16-keV beam. Not only does the position of the anticrossing shift in field amplitude, but the overall character of the interference pattern changes. Note that in the 30-GHz data the height of the first Stueckelberg oscillation is smaller than the subsequent ones.

The TWTA was the dominant source of broadband microwave noise in the system. We assume that it was predominantly shot noise, which supplies equal doses of amplitude and phase noise. Using a Hewlett Packard model 8566A spectrum analyzer and model 11970A mixer, we measured its noise power spectrum to be flat within 10 dB over the range 26.5–40 GHz. Because the TWTA noise power is essentially independent of the harmonic drive, kept well below saturation, by attenuating the TWTA output and increasing the drive we were able to vary the noise power while maintaining the same coherent microwave field amplitude in the interaction region. Figure 5 shows the experimental results for noise power densities of 0.32, 0.72, 1.34, and 2.67 mW/cm^2 corresponding to calculated rms additive field amplitudes δF of 0.8, 1.2, 1.6, and 2.3 V/cm, respectively. Clearly, the highest noise power of these four, for which δF was only about 7% of the anticrossing field amplitude, completely destroys the Stueckelberg oscillations. It also broadens the overall envelope of the signal as evidenced by the rise in signal off the baseline at fields below the location of the anticrossing. For data (not shown) that extended the amplitude range of the top scan in Fig. 5 down to $F_0=8$ V/cm, the signal was nonzero down to about 18 V/cm. Comparing the top and bottom scans in Fig. 5 in-

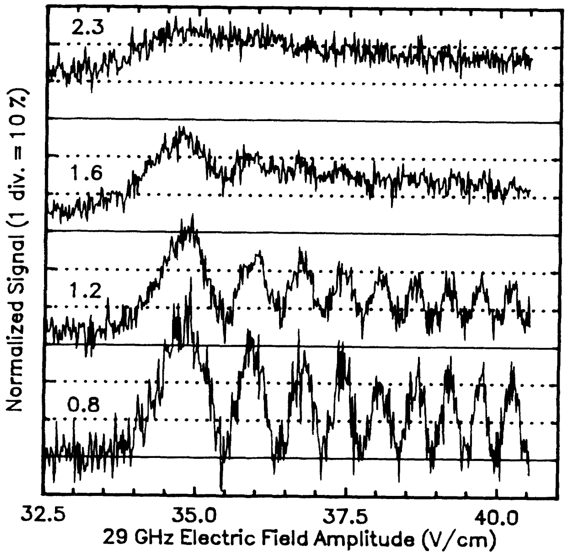

FIG. 5. 29-GHz experimental data as in Fig. 2 but for four different additive 26–40-GHz noise power densities corresponding to rms field amplitudes of 2.3, 1.6, 1.2, and 0.8 V/cm. Solid lines indicate 0% level.

dicates that the mechanism of the broadening is not likely to be an inhomogeneous one since any convolution of the bottom curve with a function whose width is of order 2.3 V/cm cannot broaden the observed nonzero signal down to $F_0 = 18$ V/cm. Thus we infer that the dominant broadening mechanism is homogeneous in nature and that noise-induced transitions to harder-to-ionize states, including but not limited to the $28\,^3P$ state, are involved.

In conclusion, our observation of Stueckelberg oscillations establishes that the physics of microwave excitation of helium Rydberg atoms can be correctly described in terms of quasistatic evolution along Floquet potential curves. In principle, the same formalism would be applicable to experiments with tightly bound atoms in focused laser pulses. The observation of Stueckelberg oscillations in pulsed laser experiments, however, would be hindered by large spatial intensity variations across the atomic sample in the focal volume, pulse-to-pulse jitter, and amplified-spontaneous-emission "noise," all of which would serve to smear out the oscillation pattern.

Our investigation of the effect of additive noise on this system directly demonstrates a point made previously [14]: Experimental observations of multiphoton transitions driven by pulsed fields cannot be understood fully without detailed knowledge not only of the atom plus field structure, but also of the precise temporal shape of the field pulse and its coherence properties (noise). Theoretically the influence of technical and thermal noise in experiments on microwave-driven Rydberg atoms was previously investigated, but this involved many coupled Floquet states [15]. Unfortunately, no numerical calculations have been performed for the much simpler present case of dynamics being dominated by a well isolated avoided crossing. This system should provide an attractive testing ground for theories seeking to incorporate the effects of noise in such problems.

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Note added.—After submission of this Letter, Baruch and Gallagher reported [16] the observation of Stueckelberg oscillations with K Rydberg atoms in a pulsed 9.3-GHz field. Their results did not show the effect of additive noise.

[a]Permanent address: Institute of Physics, Polish Academy of Sciences, Al. Lotników 32/46, 02-668 Warszawa, Poland.

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[6] Subject to certain restrictions and assumptions, the Landau-Zener formula gives an approximate expression for this transition probability.
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