Microwave Multiphoton Excitation of Helium Rydberg Atoms: The Analogy with Atomic Collisions

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We study multiphoton transitions in helium Rydberg atoms subjected to a microwave electric field of fixed frequency but varying intensity. For each principal quantum number in the range $n=25$–32, the n^3S to $n^3(L>2)$, $n=25$–32, transition probability exhibits very sharp structures as a function of the field amplitude. Their positions could be reproduced precisely using a Floquet Hamiltonian for the interaction between atom and field. Their shapes are determined by the transients of field turn-on and turn-off in a way that makes a close analogy with the theory of slow atomic collisions.

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Conventional atomic spectroscopy is concerned with the response of atoms to a radiation field whose frequency is varied. In this Letter we study, instead, excitation of helium Rydberg atoms by a microwave electric field of fixed frequency ω but varying amplitude F. For the first time we show that the associated transition probability as a function of the microwave field amplitude is marred by very sharp features for the range of principal quantum numbers we studied. Their shape is shown to be determined by the amplitude envelope function $A(t)$ that turns on, maintains, and turns off the microwave electric field, $F_z = FA(t)\cos(\omega t+\varphi)$. (The experiment averaged over initial phases φ.) The analysis of the atomic response to these transients is analogous to the well understood theory of slow atomic collisions and highlights the presence of two disparate time scales in the problem.[1] Eigenvalues of a Floquet matrix, plotted as a function of field amplitude F, play the role of potential curves in the atomic collision analog. Those potential curves are traversed slowly (compared to the sinusoidal variation of the field) when $A(t)$ rises from zero to its maximum value, and back again when the field is switched off. Because the field-switching transients are well defined, our experiments may contribute to a powerful union between atomic spectroscopy and collision physics.

Similar microwave-amplitude spectra have been reported earlier by Stoneman, Thomson, and Gallagher in the case of $n=18$–23 potassium atoms prepared in $^2S_{1/2}$ Rydberg atoms. Contrary to our findings, however, they observed only a single, very broad structure at one particular value of the principal quantum number, $n=19$.[2]

The experimental setup is sketched in Fig. 1. A beam of fast (8.95×10^5 m/s) helium atoms emerged from a gas-scattering cell in a distribution of excited states. In two separated regions of carefully tuned electric field strength, two CO_2 laser beams were used to selectively populate n^3S states. A complete account of these spectroscopic techniques was given elsewhere.[3] The n^3S Ryberg atoms next entered a microwave cavity operated

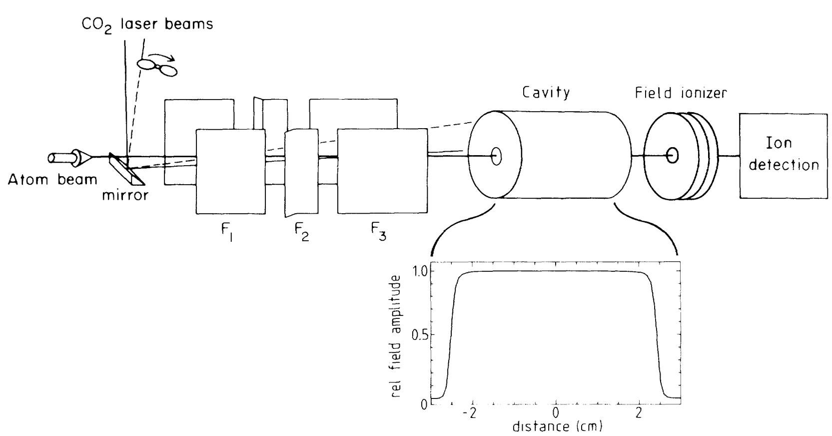

FIG. 1. Schematic drawing of the apparatus. A double-resonance laser-excitation scheme prepares helium n^3S Rydberg atoms. After exposure to about 550 oscillations of a 9.924-GHz microwave field, surviving n^3S atoms are selectively ionized. Inset: computed on-axis electric field amplitude normalized to the maximum amplitude inside the microwave cavity.

at 9.924 GHz in the TM_{020} mode, whose longitudinally directed electric field is maximal on axis. The leakage of the microwave field out of the entrance and exit holes (radius 0.129 cm) caused the gradual turn-on and turn-off of the amplitude $A(t)F$ in the rest frame of the atoms. The shape of the switching function $A(t)$ could be influenced by altering the size and shape of the holes. The radial extent of the atomic beam in the cavity was limited by a collimator (radius 0.044 cm) located just before the cavity. Figure 1 also shows the on-axis, longitudinal component F_z of the electric field amplitude, which was numerically computed using a finite-element technique.[4]

The experiment measured the fraction f of atoms that remained in the $n\,^3S$ state after interacting with the microwave field; the remainder $(1-f)$ was driven to higher angular momentum states in the same n manifold. The key experimental point was that static field ionization in the ionizer in Fig. 1 of states with azimuthal quantum number $M_L=0$ is known to be characterized by a well-defined threshold field that is about half that for $|M_L|\geq 2$.[3] Suppose that atoms were driven inside the cavity from $n\,^3S$ to $n\,^2S(L>2,\ M_L=0)$ states. The latter class would not remain in an $M_L=0$ state after leaving the cavity and traversing on their way to the ionizer a region of small, stray electric fields. Instead, the result was a wide (consistent with uniform) distribution over all states with $|M_L|<L$.[5] Thus, by setting the ionizer field just above the threshold for detecting $n\,^3S$ $(M_L=0)$ states, the loss of the $n\,^3S$-ion signal corresponded to high-L $n\,^3L$ states being produced by the microwave electric field. The ion signal was detected in phase with the chopped "F_1-laser" beam and was signal averaged in a multiscalar as a function of microwave power dissipated in the cavity. Using a procedure described in Ref. 6, measured incident power was converted to F to 5% accuracy. The signal was normalized to unit $n\,^3S$-survival probability at small power; its baseline (zero survival probability) was measured by completely quenching the $n\,^3S$ beam in a transverse electric field that was switched on briefly during each multiscalar scan.

Figure 2 shows the survival probability f as a function of the microwave electric field amplitude on the cavity (beam) axis for each n value in the range 25–32. All curves show sharp structures, in a few cases the depth of which corresponds to a microwave-induced transition probability of 0.8. In all cases the structures tend to broaden at the higher field strengths. We will show below that this phenomenon is connected to the switching transient $A(t)$ of the microwave electric field amplitude in the moving atomic rest frame. We also emphasize that the sharp structures were observed *well below* the field strength where ionization sets in, and where, necessarily, mixing with adjacent $n-1$, and $n+1$ manifolds would take place.[7]

For the case of a *constant* envelope function $A(t)$

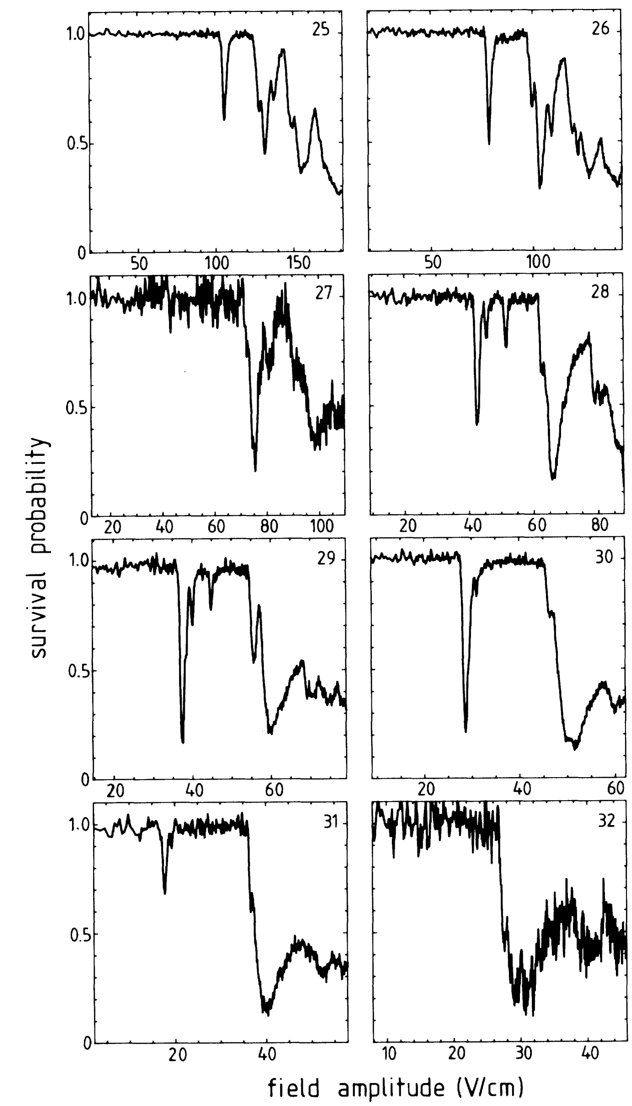

FIG. 2. Survival probability of helium $n\,^3S$ atoms, $n=25$–32, as a function of the on-axis electric field amplitude inside the microwave cavity.

[that is $A(t)=1$ for all t] the $\cos(\omega t)$ dependence of the atom-field interaction may be transformed into a time-independent semiclassical Hamiltonian by using Floquet analysis, interpreting the basis states of the Floquet matrix as direct-product atom-photon states.[8] The periodicity of the field endows the spectrum of the Hamiltonian with periodic properties. Figure 3 shows a little more than one period of a spectrum computed for $n=28$, $M_L=0$. Because we study transitions out of an ^3S state, the basis contains only triplet states that have the same parity $(-1)^{L+k}$, with k the photon number, as the initial state;[9] therefore, the periodicity in the Fig. 3 is $2\hbar\omega$ (0.620 cm^{-1}). Incidentally, the initial $28\,^3S$ state joins adiabatically to the state indicated $(L,k)=(S,8)$ [or, equivalently, $(S,10)$] in Fig. 3, whose energy bends

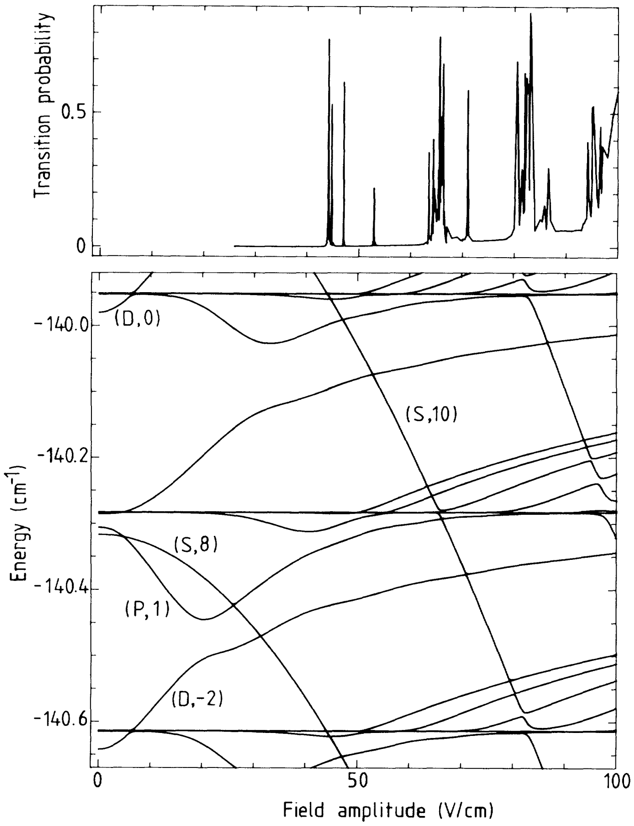

FIG. 3. Lower: map of Floquet levels of He $28\,^3S$ atoms in a 9.924-GHz microwave electric field as a function of the field amplitude. Upper: computed transition probability $28\,^3S \rightarrow 28\,^3L > 2$ using a sudden approximation and a field exposure time of 5.5×10^{-8} s.

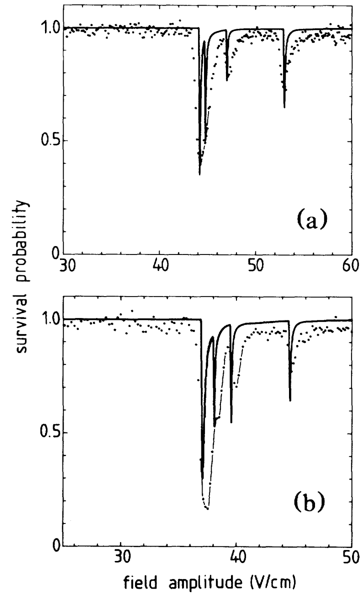

FIG. 4. (a) Dots: measured survival probability of He $28\,^3S$ atoms as a function of on-axis electric field amplitude. Full line: the complement of $n\,^3S \rightarrow n\,^3L > 2$, $n=28$ excitation probability computed from a model of six interacting states and allowing for the field-switching transients produced by the experimental $A(t)$ envelope function. (b) The same as (a), but for $n=29$.

down as the microwave amplitude increases; the energies of the final high-L states shift least with increasing amplitude. Even- and odd-parity high-L (manifold) states are spaced $\hbar\omega$ apart.

The sole remaining time dependence is that of the slowly changing amplitude envelope function $A(t)$. The turn-on of the microwave field in the rest frame of the atoms is so slow that the initial atomic $28\,^3S$ state does indeed evolve *adiabatically* into the atom-photon state denoted as $(S,10)$ and $(S,8)$ in Fig. 3. However, for field amplitudes up to 60 V/cm, the width of the well isolated anticrossings this state experiences with other states is so small that, in first approximation, they will be traversed *diabatically*. Transitions will occur when the maximum field amplitude in the cavity reaches out to and lingers at each of those anticrossings. The transition probability $28\,^3S \rightarrow 28\,^3L$, incoherently summed over all $L > 2$, is also shown in Fig. 3. It was computed using the adiabatic continuation of the bare atomic $28\,^3S$ state as initial state and taking the traversal time through the cavity (5.5×10^{-8} s) as the interaction time with the field switched suddenly (with respect to the inverse anticrossing widths). Sharp peaks in the transition probability are predicted precisely (within the calibration uncertainty of the electric field strength) where the experimental curves have sharp dips.[10] The experimental and theoretical curves have *not* been normalized in field strength; the comparison is absolute.

In order to understand the *shape* of the observed dips, we must realize that our assumption of diabatic traversal of anticrossings does not hold when the maximum field amplitude inside the cavity was set right at an anticrossing and the rate of change of the level separation vanished. When the maximum field strength was increased slightly, the anticrossing was traversed twice, once at the cavity entrance and again at its exit. Consequently, the transition probability should exhibit interferences analogous to the well-known Stückelberg oscillations in atomic collision theory. At each anticrossing the multiphoton transition probability is given by the Landau-Zener-Stückelberg formula.[1,11,12] The broadening of the structures at the higher amplitude in Fig. 2 can now be understood as partial adiabatic and partial diabatic passage of anticrossings.[13]

Figure 4 shows a comparison between measured and computed spectra for $n=28$ and 29. Computations were based on an integration of the time-dependent Schrödinger equation in the slowly varying envelope function $A(t)$. An adequate model of the six most relevant interacting states was extracted from Fig. 3, with their pairwise interaction matrix elements deduced from the graphed anticrossings widths. The energies of the model states were also taken to be linearly dependent on the field amplitude. We averaged the resulting spectra over the 0.2% radial variation of the experimental electric field amplitude. The calculated spectra do exhibit tails at the high-field side of the dips, tails that are also observed in the experiment. These contain unresolved Stückelberg oscillations which may be able to be resolved in future experiments using a very carefully designed envelope function $A(t)$. An important present discrepancy, however, is that the calculated features are significantly narrower than those observed in the experiment. This may be related to properties of the radiation field that affect the coherence and need to be modeled.[13]

The phenomena discussed in this Letter are a beautiful illustration of strong-field effects in multiphoton excitation of atoms and molecules. Precise control over the amplitude envelope function $A(t)$ permits detailed study of important transient effects of a kind that will be much harder to control in experiments using lasers.

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[8]J. H. Shirley, Phys. Rev. **138**, B979 (1965); K. F. Milfield and R. E. Wyatt, Phys. Rev. A **27**, 72 (1983).

[9]A. Maquet, Shih-I Chu, and W. P. Reinhardt, Phys. Rev. A **27**, 2946 (1983). Strictly speaking k is a Fourier index which may, however, be interpreted as a relative photon number in the semiclassical limit (see Shirley, Ref. 8).

[10]Stoneman, Thomson, and Gallagher (Ref. 2) have earlier sought an explanation of their K microwave excitation experiments along the same lines. However, Ref. 2 gives no consideration to transition probabilities or transients. From Fig. 3 the importance of the first may be gleaned: Not all avoided crossings in the lower part of Fig. 3 give rise to sizable transition probabilities in the upper part of Fig. 3. This is particularly clear for the case of $n=26$, where the calculation (not shown) predicts avoided crossings of the 26^3S state with $26^3L>2$ states at 46 V/cm and again at 77 V/cm, but where the first structure is not observed until 78 V/cm.

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[12]Note that the Landau-Zener-Stückelberg formula is applicable only when the anticrossings are well separated. This is not the case when the maximum field strength inside the cavity is set precisely at the anticrossing, and, instead, the so-called Nikitin exponential model applies (see Ref. 1).

[13]W. van der Water *et al.* (to be published).