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Rabi-nutations were observed for four- and six-photon microwave transitions between the Rb $66P_{3/2}$ state on the one hand and the $68P_{3/2}$ and $69P_{3/2}$ states on the other hand, respectively. The applied frequencies for the four- and six-photon transitions were 12348.1 MHz and 12071.7 MHz respectively. The measured Rabi-frequencies agree with the theoretically expected power dependence. For the four-photon transitions, the transition probability was calculated by means of perturbation theory. The agreement with the experiment is fairly good.

1. Introduction

In this paper, we report on the first observation of four- and six-photon Rabi-nutations between P-states of rubidium Rydberg atoms in an external microwave field. Our experimental setup (see fig. 1) was built to investigate chaos phenomena of Rydberg atoms in strong microwave fields. A brief description of the apparatus has been published in ref. [1] together with initial experimental results. The measurements of the Rabi-nutations described in this paper have been performed as a test of the setup.

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Fig. 1. Scheme of the experimental setup. The timing of the laser light pulse, the microwave pulse and the field ramp is controlled by the pulse generator and delay lines.

Rabi-nutations are of interest for this purpose since they allow to check the accurate timing of the microwave field pulses inducing the transitions. Furthermore, they allow to calibrate the strength of the field. For the latter purpose, however, the transition matrix elements have to be known. To calculate those is usually rather accurately possible for single-photon or two-photon transitions. When it comes to higher order multi-photon transitions, the field strength is so high that the ac Stark-effect may play an important role. The measurements of the Rabi-nutations are then a good test for the calculations of the multi-photon matrix elements. It should also be mentioned here that in a recent publication, Gentile et al. [2] reported on measurements of the Rabi-nutations of one- and two-photon transitions between Rydberg states of calcium. The experimental setup used for those measurements is similar to the one used by us.

2. Experimental setup

Rubidium atoms of an atomic beam are excited to the $66P_{3/2}$ state by a short linearly polarized laser pulse (duration 6 μs). The excitation pulse is generated by switching the beam of a frequency-doubled cw dye laser by means of an electrooptical modulator. The resulting group of excited atoms then en-

ters a rectangular waveguide with a cross section of $0.95 \times 1.90 \text{ cm}^2$. Due to the thermal velocity distribution, the group of excited atoms slightly expands geometrically. However, the duration of the laser pulse is adjusted so that the spread of the atoms stays smaller than the width of the waveguide (0.95 cm) during the whole interaction time with the microwave field. This is switched on after all atoms entered the waveguide. The mode of the microwave field is TE_{01} with the direction of the electric field parallel to the polarization of the laser light. The duration of the pulse can be varied in steps of 0.5 ns from 5 to 500 ns. Since all atoms have the same interaction time with the microwave field, their final state distribution changes periodically with the duration of the microwave pulse.

After the interaction with the microwave field, the rubidium atoms reach the ionization region. Here, they are ionized in an electric field the strength of which is changed linearly in time after the total group of atoms entered between the capacitor plates (slew rate $1 \text{ V/cm } \mu\text{s}$). The ejected electrons are detected by a channeltron multiplier. Owing to the field varying in time, a state-selective detection of the Rydberg atoms is possible: the highly excited states are ionized at smaller field strengths than the lower-lying ones. For the observation of the Rabi nutations, either the upper or the lower state atoms have to be detected; this is done in our experiment by counting the field electrons during a suitable time window of the field sweep.

The microwave pulse induces four- or six-photon transitions depending on the microwave frequency. In the experiment, either a 1720 Systron-Donner Synthesizer or, alternatively, a stabilized Marconi Micro Sweep Oscillator type 6600A were used as microwave generators. For both oscillators, the output power at the first harmonic was 0 dBm whereas the power at higher harmonics was less than -50 dBm . The microwave frequencies used were 12348.1 MHz for the observation of the four-photon Rabi-nutations of the transition $66\text{P}_{3/2}-68\text{P}_{3/2}$ and 12071.7 MHz for the six-photon Rabi-nutations of the transition $66\text{P}_{3/2}-69\text{P}_{3/2}$. It should be mentioned here that recently we have also observed the two-photon Rabi-nutations [3], e.g. for the $67\text{P}_{3/2}-68\text{P}_{3/2}$ transition. The resulting Rabi-frequency was used to calibrate the microwave field strength in the wave-

guide. This accurate calibration is necessary for the chaos experiment [1].

3. Results

An example of the four-photon Rabi-nutation signals (transition $66\text{P}_{3/2}-68\text{P}_{3/2}$) is shown in fig. 2 for different microwave powers: $3.24 \times 10^{-5} \text{ W}$ (lower curve) and $3.98 \times 10^{-5} \text{ W}$ (upper curve). The curves show the population of the upper levels versus time duration of the microwave pulse. The corresponding Rabi-frequencies are $(6.32 \pm 0.10) \text{ MHz}$ and $(9.12 \pm 0.25) \text{ MHz}$ for the lower and upper signals, respectively. These two Rabi-frequencies are plotted versus the square of the microwave power in fig. 3 together with three other measurements. As expected from theory, the results of all measurements lie on a straight line.

Due to the ac Stark-effect, the levels shift with increasing power of the microwave field. Since the Rabi-frequency depends on the detuning of the lev-

Fig. 2. Four-photon Rabi-nutations for the transition $66\text{P}_{3/2}-68\text{P}_{3/2}$. Microwave powers $3.98 \times 10^{-5} \text{ W}$ (lower curve).

Fig. 3. Rabi-frequency for the four-photon transition $66\text{P}_{3/2}-68\text{P}_{3/2}$ versus the square of the microwave power.

els, the ac Stark-shift may cause an additional change in the Rabi-nutation. To compensate for that the accurate four-photon resonance frequency was determined at each power level and after that the Rabi-frequency was measured.

It is interesting to compare our results with theoretical predictions. The multi-photon transition probability can be calculated using higher order perturbation theory. The matrix element for a K -photon transition between the levels $|n\rangle$ and $|k\rangle$ are given by [4]

$$V_{kn}^{(K)} = \frac{1}{2^K h^{K-1}} \times \sum_{\text{virtual states } m, s, \dots, p} \frac{V_{km}^{(1)} V_{ms}^{(1)} \dots V_{pn}^{(1)}}{[\nu_{mn} - (K-1)\nu] \times \dots \times [\nu_{pn} - \nu]}$$

where ν is the microwave frequency, $V_{km}^{(1)}$, $V_{ms}^{(1)}$, ... are one-photon matrix elements and ν_{km} , ν_{ms} the corresponding frequency differences. The connection between the Rabi-frequency and V_{kn} is given by

$$\nu_R = (2/h) F^K V_{kn}^{(K)},$$

where F is the amplitude of the microwave field. The formula is only valid for zero detuning.

In order to calculate $V_{kn}^{(4)}$, the states between $n=64$

Fig. 4. Six-photon Rabi-nutations for the transition $66\text{P}_{3/2}-69\text{P}_{3/2}$. Microwave power $2.51 \times 10^{-4} \text{ W}$ (upper curve) and $2.24 \times 10^{-4} \text{ W}$ (lower curve).

Fig. 5. Rabi-frequency for the six-photon transition $66\text{P}_{3/2}-69\text{P}_{3/2}$ versus the third power of the microwave power.

and 70 and l between 0 and 3 were taken into account. The respective dipole matrix elements were calculated using the method of Oumarou et al. [5]. The result of the calculation is

$$(2/h)V_{kn}^{(4)} = 4.942 \text{ MHz}/(10^4 \text{ V}^4/\text{m}^4) .$$

The corresponding experimental value which follows from fig. 3 is $(4.29 \pm 0.07) \text{ MHz}/(10^4 \text{ V}^4/\text{m}^4)$. The error follows from the uncertainties of the measurements plotted in fig. 3. To extract the result from fig. 3, the value for the microwave power had to be converted into the microwave field amplitude. The power was measured using a precision power meter. To calibrate the field, the two-photon transition $66\text{P}_{3/2} - 67\text{P}_{3/2}$ has been used. The frequency applied to measure this transition was 12634.85 MHz. This two-photon frequency is the closest one to 12348.1 MHz which had to be used for the four-photon transition. The factor between the square of the power and the fourth power of the microwave field amplitude resulting from the calibration measurements is $F^4/P^2 = 13.11 (10^2 \text{ V}^4/\text{m}^4 \text{ W}^2)$. Repeated measurements at different days show a variation of this value of about $\pm 3\%$. The calibration factor may change due to the fact that the waveguide slightly gets coated by Rb-atoms. This results in a reduction of the field amplitude of the microwave signal. Therefore it is most likely that this variation of the calibration factor does explain the major part of the difference between theory and experiment for the matrix element of the four-photon transition.

We also succeeded in measuring six-photon Rabi-

nutations. Two results for those measurements are shown in fig. 4. The power was $2.51 \times 10^{-4} \text{ W}$ for the upper curve and $2.24 \times 10^{-4} \text{ W}$ for the lower curve. Due to the ac Stark-effect, the resonance frequency had to be readjusted after the power of the microwave field was changed. At the two power levels the transition frequencies were 12071.24 MHz (upper curve) and 12071.7 MHz (lower curve). The difference corresponds to the variation of the ac Stark-effect between the two power levels.

The Rabi-frequencies which result from the measurements displayed in fig. 4 are $(5.67 \pm 0.11) \text{ MHz}$ (upper curve) and $(3.95 \pm 0.07) \text{ MHz}$ (lower curve). These values are plotted in fig. 5 versus the third power of the microwave power together with the results of three other measurements. Again, as expected from theory, the results lie on a straight line. In the case of the six-photon transitions, no comparison with theory was performed.

References

- [1] R. Blümel, R. Graham, L. Sirko, U. Smilansky, H. Walther and K. Yamada, *Phys. Rev. Lett.* 62 (1989) 341.
- [2] T.R. Gentile, B.J. Hughey, D. Kleppner and T.W. Ducas, *Phys. Rev. A* 40 (1989) 5103.
- [3] R. Blümel, R. Graham, L. Sirko, U. Smilansky, H. Walther and K. Yamada, in: *Fundamentals of laser interactions II*, ed. F. Ehlotzky, *Lecture Notes in Physics*, Vol. 339 (Springer, Berlin, 1989) p. 218.
- [4] N.B. Delone and V.P. Krainov, *Atoms in strong light fields* (Springer, Berlin, 1985).
- [5] B. Oumarou, J. Picart, N. Tran Minh and J. Chapelle, *Phys. Rev. A* 37 (1985) 1885.