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Abstract. – We present first experimental results on the interaction of rubidium Rydberg atoms with bichromatic microwave fields. The experiments are performed in a regime where the effect of localization of the population distribution is observed for purely monochromatic fields. The second frequency leads to a significant destabilization of the population distribution. The experimental observations are compared to theoretical results.

During the past years much attention has been devoted to theoretical and experimental investigations of highly excited hydrogen atoms in intense microwave fields [1-5]. This interest is due to the fact that hydrogen atoms in microwave fields represent a good example of a nonlinear and classically chaotic system. Within a certain range of field strength, field frequency and initial value of the principal quantum number, however, the quantum-mechanical treatment shows that the number of states contributing to the atomic dynamics is limited, despite the fact that the corresponding classical counterpart shows diffusion in the action variable [1]. This higher stability of quantum dynamics, contrasting classical dynamics, led to several approaches for the theoretical interpretation [1, 3, 6, 7]. For the present paper we concentrate on the frequency range where dynamical localization is supposed to occur and which was investigated in several experiments recently [4, 5, 7]. It seems that there is no doubt any more on the existence of this phenomenon as a quantum-mechanical interference effect and the natural next step is to isolate sufficient and necessary conditions for its appearance, in theory as well as in numerical and in laboratory experiments. In other words the question arises under which conditions one can destroy the quantum localization experienced by a hydrogen atom in a monochromatic coherent electromagnetic field. Assuming that periodicity of the Hamiltonian—corresponding to the applicability of the Floquet picture—is a typical property of nonlinear systems which display dynamical localization one expects that broadband noise, which spoils periodicity, destroys this latter effect. Indeed, this has been shown in recent theoretical and experimental work [7].

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Another approach for testing the stability of localization is to introduce a second discrete frequency, instead of broadband noise. This allows—in principle—to control the periodicity properties of the Hamiltonian by proper choice of the second frequency. In this paper we present measurements demonstrating the strongly destabilizing influence of a second coherent field component on the localization properties of rubidium Rydberg atoms. These first experiments in the frequency range where dynamical localization is supposed to occur [1] show that this destabilization is much more efficient than in the low-frequency regime covered by the experiments performed by Moorman *et al.* [8] and numerically studied by Blümel and coworkers in ref. [9] and [10].

Our experimental approach is in principle identical to that of a recent study of the dynamical localization of rubidium Rydberg atoms in a monochromatic field and of the stability of this phenomenon with respect to broad-band noise. Details of the experimental set-up will be published in ref. [7]. We only should like to recall here that the experimental geometry allows for a well-defined microwave interaction time being continuously adjustable from roughly 10 ns to about 20 μ s. The atoms experience a microwave pulse with a flat top, the switch-on and -off time of which is close to 6 ns. In the two-frequency experiment, a 1720 Systron-Donner Synthesizer and a stabilized Marconi Micro Sweep Oscillator type 6600 A have been used as microwave generators. Both oscillators produced a microwave signal which was practically free of higher harmonics (at least 50 dB below the carrier frequency). The phases of the two microwaves were uncorrelated and averaged over during data taking. However, due to the large coherence time of both microwave synthesizers, the phases ϕ_1 and ϕ_2 can be regarded as constant during one microwave pulse. Since an accurate determination of the field amplitude experienced by the atoms is necessary, we confined ourselves to microwave frequencies which correspond to two-photon resonances between $p_{3/2}$ states of principal quantum number much lower than that of $84p_{3/2}$ used as initial state for the two-frequency measurements. The field amplitude was then determined using the Rabi-frequency of the two-photon transition. This method of calibration has been described in detail in a recent publication [11].

In the experiments described here, we investigated the population redistribution of Rydberg atoms under the influence of external bichromatic microwave fields, as a function of the duration of the microwave interaction. The behaviour induced by the bichromatic field was compared to the effect of a monochromatic field. In the bichromatic case, we chose the field amplitude of each component to be equal to each other (0.27 V/cm), the sum of both equalizing the value of the amplitude in the monochromatic case (0.54 V/cm). Consequently, in the bichromatic case, the total power was only half as large as in the monochromatic case. The microwave frequencies chosen in this experiment were 9650.40 MHz, 10 077.39 MHz and 12 059.44 MHz. In fig. 2a-f) we show the static-field ionization probability $P(E, t)$ for the interaction with a monochromatic field, as a function of the ramp field E and for different interaction times $t = 10$ ns, 100 ns and 1 μ s. Comparing these graphs to fig. 1, which represents the field ionization probability $P(E, t = 0)$ of the initially prepared state $84p_{3/2}$ for no microwave interaction, we recognize that the microwave frequency 12 059.44 MHz leads to a much stronger (and nontrivial) level redistribution [7] than the frequency 9650.40 MHz. The application of 10 077.39 MHz induces an effect similar to the one observed for 9650.40 MHz and is therefore not shown here. This observation clearly demonstrates a different effectiveness of the frequencies used since the field amplitude has been fixed at the same value in all three cases. As we want to compare the time evolution of the level redistribution in the presence of mono- and bichromatic fields, respectively, we plot the relative, time-dependent change of the width of the population distribution of the atoms with respect to the distribution after an interaction time of 10 ns, for each frequency combination. A convenient measure for this spreading behaviour is the χ^2 -function, de-

Fig. 1. – Field ionization probability $P(E, t=0)$ of the initial state $84p_{3/2}$, without microwave interaction. P is normalized to the integral count rate of field-ionized atoms. The value of this integral was close to 25 000 for all the scans displayed in fig. 1-3. The double-peak structure of the ionization signal is due to adiabatic and diabatic ionization, respectively.

defined as

$$\chi^2(t) \sim \int \left\{ \frac{P(E, t) - P(E, 10 \text{ ns})}{P(E, t)^{1/2}} \right\}^2 dE, \quad t > 10 \text{ ns}. \quad (1)$$

In fig. 3 the field ionization probabilities $P(E, t)$ for a bichromatic field made up by the frequencies 9650.40 MHz and 12 059.44 MHz are shown. As immediately evident, there is a quite effective redistribution of the level population already after 10 ns interaction time. For 1 μ s, the distribution is rather broadened even compared to fig. 2f), representing the largest effect of the monochromatic field observed for the given parameters. A similar result has been obtained for the frequency pair 10 077.39 MHz and 12 059.44 MHz. For all the configurations represented in fig. 2 and 3 χ^2 is plotted in fig. 4. Whereas, for the monochromatic fields, χ^2 shows an almost constant behaviour, the presence of a second field component leads to a very fast, effective and unlimited spreading, the difference in the time for the beginning of the delocalization processes being only barely resolvable. The results show clearly that the bichromatic fields strongly destabilize the atomic dynamics—at a total power of the field only half as large as in the monochromatic case!

Fig. 2. – Field ionization probabilities (normalization as in fig. 1) after a microwave interaction of 10 ns, 100 ns and 1 μ s duration (columns from left to right); for the microwave frequencies 9650.40 MHz (a-c)) and 12 059.44 MHz (d-f)), respectively. Initial state (microwave amplitude) in all cases: $84p_{3/2}$ (0.54 V/cm).

Fig. 3. - Field ionization probabilities (normalization as in fig. 1) after a microwave interaction of a duration of *a*) 10 ns, *b*) 100 ns and *c*) 1 μ s; for a bichromatic microwave field with frequencies 9650.40 MHz and 12 059.44 MHz. Amplitude of each field component: 0.27 V/cm.

Fig. 4. - Change of the width of the population distribution in time, measured by χ^2 defined in (1). χ^2 is plotted in arbitrary units. Each point in the figure corresponds to the average of at least four individual measurements. Monochromatic microwave fields with an amplitude of 0.54 V/cm each: pyramids: 9650.40 MHz, crosses: 10 077.39 MHz, circles: 12 059.44 MHz. Bichromatic microwave fields with an amplitude of 0.27 V/cm of each component: triangles: 9650.40 MHz and 12 059.44 MHz; diamonds: 10 077.39 MHz and 12 059.44 MHz.

In the following we should like to compare our results with those of other groups (mainly using hydrogen atoms) as well as with the concept of localization and delocalization. For the sake of comparison we should discuss our findings in terms of scaled frequency and scaled field amplitudes. However, since we are using rubidium instead of hydrogen, we have to account for the lack of angular-momentum degeneracy in our case which leads to a much more complicated level structure than in the hydrogen experiments. It is well known that the r^{-1} -potential is very important for the classical scaling properties as well as for the classical chaos border [12]; therefore it cannot be excluded *a priori* that core effects may be of importance for the dynamics of the state distribution of rubidium in the classical as well as in the quantum-mechanical description. Since there are no rigorous classical-model calculations available for rubidium atoms, we shall try to discuss our results on a semi-empirical basis. A detailed discussion of the problem of scaling will be provided elsewhere. We start with a rough argument for an appropriate definition of the scaled frequency. Due to the typical level spacing of rubidium being of the order of half the hydrogenic spacing, we can assume that mainly levels with $l \leq 3$ will dominate the multiphoton process we are dealing with. Consequently, defining the scaled frequency ω_0 as the ratio of microwave frequency and typical level spacing, we argue that for rubidium atoms ω_0 should be at least twice as large as for hydrogen atoms. With the definition

$$\omega_0 := 2 \cdot \omega \cdot n_0^3 \quad (2)$$

the frequencies used in the experiments described above are then 1.74 ($\equiv 9650.40$ MHz), 1.82 ($\equiv 10\,077.39$ MHz) and 2.18 ($\equiv 12\,059.44$ MHz). For the scaling of the field strength, there is no such simple argument. From the discussion in the previous section it is clear that our experiments have been performed for values of the scaled frequency larger than one. Up to now there are no theoretical calculations available in this regime except a paper by Leopolds and Richard who consider their results as preliminary [13]. It is interesting to mention the qualitative consistency of our results with predictions by Casati *et al.*, published in a recent preprint [14]. These authors found—as a result of numerical simulations for hydrogen with $\omega_0^H = \omega \cdot n_0^3 > 1$, *i.e.* in the high-frequency domain—the delocalization border to be decreased by a second frequency of the microwave field. Similarly, also in ref. [13] the number of accessible states has been observed to be increased by a second frequency, nonetheless still remaining smaller than in a classical model. However, the experimental result shows an even higher than predicted efficiency of the second field component (see, *e.g.*, table II in ref. [14]), and a qualitative comparison is not straightforward because of the relatively short interaction time of less than 300 field oscillations used in the numerical simulations (in our experiments we have about $100 \div 10\,000$ field cycles). In addition, there is the problem of amplitude scaling mentioned above. Furthermore, the authors of ref. [13] used a sinusoidal envelope of the microwave pulse—a condition which does not apply for our set-up and which might be crucial for comparison [3, 13].

Another important point is that Casati and coworkers observed an enhancement of delocalization only for two incommensurate frequencies and at first glance it is not clear how to transfer terms like commensurability and incommensurability to real experiments dealing with finite spectral widths and finite interaction times. Blümel *et al.* [9, 10] have been able to reconstruct experimental data produced by Moorman *et al.* [8] by approximating the frequency ratio used in these experiments by continued-fractions and then using Floquet theory. The proper continued-fraction approximand was determined by the condition that the difference between the real ratio and the approximand had to be less than the experimental resolution of this ratio, determined by the frequency spread corresponding to the microwave interaction time. For our experiments and for the maximal interaction time

of 1 μ s, the corresponding approximands are 677/846 for the pair 9650.40 MHz and 12 059.44 MHz, and 61/73 for the pair 10 077.39 MHz and 12 059.44 MHz, respectively.

However, since we are resolving the time dependence of χ^2 , it should be an interesting question whether the commensurate or incommensurate character of the frequency ratio—basically determined by its continued-fraction expansion—does emerge in a fine-scaled structure of $\chi^2(t)$, as a result of the frequency-time uncertainty reducing the bandwidth of the applied microwaves as a function of time. However, to investigate this problem averaging of the phases of the microwaves should be prevented and more suitable frequency pairs should be chosen to begin with.

To finish this paper, we should like to stress an important difference of the results of Moorman *et al.* [8] for $\omega_0 < 1$ and of our data. As pointed out in Blümel's [9, 10] analysis of Moorman's data, in the low-frequency regime the maximum value of the field—the sum of the amplitude of the two components in the bichromatic case—is the relevant parameter for the delocalization threshold, i.e. for the onset of ionization. (Results recently published by Ruff *et al.* [15], who exposed Na atoms to low-frequency bichromatic fields, support this conclusion.) Blümel's calculations were based on the investigation of the behaviour of a width function, which measures the width of an atomic bound state in the Floquet basis and which exhibits a sharp transition from small to large values [2] at field parameters which were then chosen to define the ionization threshold. As far as this definition is still appropriate for the regime $\omega_0 > 1$, where (for a monochromatic field) the transition behaviour of the width function has been found to be smoother than in the low-frequency domain, our results show a qualitatively different influence of a second microwave frequency in the high-frequency regime. Since χ^2 is an immediate experimental measure of the width function, it is evident that here the same value of the field amplitude yields a drastic delocalization in the bichromatic case when the monochromatic excitation leads to a well-localized behaviour. This should then show up as a significant decrease of the ionization threshold in ionization experiments.

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