Experimental Study of Quantum and Classical Limits in Microwave Ionization of Rubidium Rydberg Atoms

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First experiments are described on the impact of additive noise on the ionization of Rb Rydberg atoms in microwave fields. Dynamical localization and its gradual destruction by noise are observed. The threshold and time dependence of the ionization process are investigated in the presence as well as in the absence of noise. Purely stochastic excitation induces a diffusionlike behavior which might be interpreted as restoration of the classical limit.

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The study of the time evolution of a quantum system whose classical counterpart exhibits chaotic dynamics has received a great deal of attention in recent years. One of the most prominent features of some of these systems, known as "dynamical localization," is that their quantum evolution follows the diffusive classical dynamics only initially, whereas after a certain break time, quantummechanical coherence effects can change the dynamics considerably [1]. Experiments performed to test this striking phenomenon investigated the population change and the ionization process of Rydberg atoms induced by strong microwave fields [2-4], and there have been found several approaches of theoretical interpretation [3-7]. In several papers it has been discussed that the presence of noise can change significantly the dynamics of the excitation process [4,8-10]. In this Letter, we report on the first experimental studies of the ionization process of rubidium Rydberg atoms in the presence of monochromatic and/or stochastic microwave fields. We show that the presence of a small amount of noise decreases the ionization thresholds in the regime of localization. Furthermore, the functional form of the quantum diffusion coefficient D_q describing the ionization process could be determined for the first time.

Our experimental setup is basically identical to the one described in Ref. [4]. We only recall here that the experimental geometry allows for a well-defined microwave interaction time being continuously adjustable from roughly 10 ns to about 10 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. The coherent microwave source used was a Systron-Donner Model 1720, 0.05-18-GHz synthesizer which delivers a microwave signal of high spectral purity (higher harmonics and broadband noise are at least 55 dB below the carrier; the linewidth of the continuous microwave signal is smaller than 30 Hz). The noise source is a traveling-wave-tube amplifier for the frequency range between 8 and 18 GHz (variation of the output signal in the plateau of the amplifier characteristic is below 10 dB) which can be considered as an emitter of white noise. The linear polarization vector of the microwave radiation lies parallel to the atomic beam direction. There is a cutoff value n_c for the principal quantum number above which the atoms are ionized by stray or local electric fields in our apparatus; n_c was estimated to lie beyond 135 for the present experiments. The atoms surviving in Rydberg states are detected by ionization in a static electric field. The ionization probability of the atoms in the microwave field is then measured as follows: First, we count the number of atoms initially laser excited to Rydberg states, in the absence of any microwave signal (S_{off}) . Immediately after that the next measuring cycle, with a microwave signal being applied, is started and the number of atoms that survive the presence of the microwave pulse (S_{on}) is counted. From these count rates we extract the ionization probability as $P = [S_{off}]$ $-S_{on}]/S_{off}$. The ionization signal includes atoms excited to the continuum as well as atoms which are excited to principal quantum numbers higher than n_c . The strength of the microwave field experienced by the atoms was calibrated using two-photon Rabi nutations [11]. The noise contribution to the signal was determined by use of an average calibration factor gained from the results for monochromatic fields. In order to keep thermal radiation as small as possible we performed our experiments at liquid-nitrogen temperature.

In our experiments we measured the ionization probability P of rubidium Rydberg atoms which were subjected to (a) a coherent microwave signal (v=8867 MHz), (b) a monochromatic field plus added broadband noise (bandwidth 8-18 GHz), or (c) a pure broadband noise signal. The ionization probability is detected as a function of the microwave field amplitude ε_c [cases (a) and (b)] or as a function of the root-mean-square value of the average microwave power ε_N [case (c)]. In all cases we determined the 10% ionization threshold fields $\varepsilon_{10\%}$ from the experimental results.

In the experiments on the ionization probability of rubidium Rydberg atoms in an intense monochromatic field plus a broadband microwave noise the integral power of the stochastic component, when present, was always at least 20 dB below the power of the monochromatic field.

The atoms were prepared in an initial Rydberg state, with principal quantum numbers n_0 between 55 and 95 for the pure monochromatic case and between 60 and 91 when broadband noise was added to the microwave signal. For all measurements the monochromatic signal had a frequency of 8867 MHz and the interaction time was fixed to 5 μ s. In Fig. 1 the normalized ionization thresholds $\varepsilon_{0,10\%} = \varepsilon_c (10\%) n_{0,\text{eff}}^4$ are plotted as a function of the frequency $(n_{0,eff}$ is the effective quantum number). For comparison with theory, usually the scaled frequency ω_0 is used, corresponding to the ratio between the applied microwave frequency and the Kepler frequency, which can be easily calculated in the case of the hydrogen spectrum. In the case of an alkaline spectrum where we have a large *l* splitting we have to pay attention to the problem of which frequency we use for normalization. We have plotted two alternative ways of normalization which correspond to the two nearest dipole-allowed upward transitions starting from a p state, being $np \rightarrow (n+1)s$ and $np \rightarrow (n-1)d$. This semiempirical definition of the scaled frequency for rubidium, which gives qualitatively the same results as an averaging argument used in earlier work (see, e.g., Ref. [4]), is useful for a qualitative comparison with results obtained by hydrogen experiments and appropriate from the viewpoint of atomic physics. However, it should be clearly distinguished from the no-



FIG. 1. Normalized threshold field for 10% ionization, $\varepsilon_{0,10\%}$, as a function of the normalized frequency. $\varepsilon_{0,10\%} = \varepsilon_c (10\%)$ $\times n_{0,eff}^4$; $n_{0,eff} = n_0 - \delta$, where $\delta = 2.64$ is the quantum defect of the initial p state [4]. Two different transitions have been used to normalize the frequency: $\omega_{ps} = 2 \times 8867 \text{ MHz}/2Rn_{0,eff}^{-3}$ for the transition $np \rightarrow (n+1)s$ (upper scale) and alternatively $\omega_{pd} = (3.4 \times 8867 \text{ MHz})/2Rn_{0,eff}^{-3}$ for the transition $np \rightarrow (n - 1)d$ (lower scale). R denotes the Rydberg constant in MHz. The solid line connects the values measured for monochromatic excitation. The points represent threshold fields observed in the presence of additive noise. We used four different levels of integrated noise power: $\times, -35$ dBm; $\Delta, -25$ dBm; $\Diamond, -20$ dBm; and +, -15 dBm. The statistical and systematic errors of $\varepsilon_{0,10\%}$ are estimated to be 5% each.

tion of the scaling properties of the classical Hamiltonian which arise as a consequence of the Coulomb potential present in the hydrogen problem and which are no longer exact for the non-Coulombic core potentials of alkaline atoms. For the monochromatic case (solid line) there is an increase of the ionization threshold as a function of the scaled frequency, showing the localization phenomenon. With an increasing noise level the resulting ionization field strength shows a smaller slope which finally gets negative. The influence of the additive noise increases when we increase the value of the initial principal quantum number n_0 . For each value of the initial quantum number the noise levels used were -35, -25, -20, and -15 dBm. We interpret these experimental observations in terms of destruction of the coherence properties of the quantum system due to the presence of additive noise; the system then approaches the classical diffusion process.

In the region $n_0 > 70$ where the phenomenon of localization is observed we performed an experimental study of the dependence of the ionization threshold on the microwave pulse length. We use $\varepsilon_{10\%}$ to denote $\varepsilon_c(10\%)$ in the monochromatic case and $\varepsilon_N(10\%)$ in the case of excitation by pure noise. In Fig. 2 $\varepsilon_{10\%}$ is plotted as a function of time, with $n_0 = 85$, for both coherent and stochastic excitation. From this fully logarithmic plot it is apparent that in both cases the data are in agreement with a power-law behavior $\varepsilon_{10\%} \propto \tau^{-\alpha}$ in a wide interval. Least-squares fits to the data result in $\alpha = 0.13 \pm 0.04$ (monochromatic case) and $\alpha = 0.48 \pm 0.04$ (noise case). The intervals where the fits were performed are 30 ns-10 μ s for the coherent case and 30 ns-1 μ s for the stochastic case. We repeated the experiments for the coherent case with different values of n_0 . Again we observed a behavior according to $\varepsilon_{10\%} \propto \tau^{-\alpha}$, and the results obtained for α



FIG. 2. Time dependence of the 10% ionization threshold fields $\varepsilon_{10\%}$ for coherent signal excitation, $\varepsilon_{c,10\%}$, and for broadband noise excitation, $\varepsilon_{N,10\%}$. The initial principal quantum number was $n_0 = 85$ and the frequency 8867 MHz. The errors of $\log_{10}[\varepsilon_{10\%}/(V/m)]$, here and in Fig. 3, are of the order of the size of the symbols used to identify the measurements.

TABLE I. Values for α for different initial quantum numbers n_0 (monochromatic excitation $\nu = 8867$ MHz). The two results given for $n_0 = 84$ have been obtained at different stages of the experiment. Their difference can be considered as a measure for the reproducibility of the determination of α .

<i>n</i> ₀	a	
75	0.25 ± 0.06	
84	0.20 ± 0.04	
84	0.27 ± 0.04	
85	0.13 ± 0.04	
95	0.20 ± 0.04	

are collected in Table I. The measured values depend on the initial principal quantum number and range between 0.13 and 0.27.

From our experimental observation we can extract the functional form of the quantum diffusion coefficient D_q for excitation by noise only. The system reaches a defined degree of ionization when the product $D_q \tau$ equals a given constant value K which defines the amount of ionization (10% in our case). We tested the results for D_q according to $D_q \propto \varepsilon^{\kappa}$, found for the quantum kicked rotator subjected to a broadband external noise signal [8,10]. The theory predicts $\kappa = 2$. Our experimental result is $\kappa = 2.1 \pm 0.1$. It is worth noting that although the theoretical study was performed for the kicked rotator, the results are expected to be model independent [8]. Moreover, the functional form of the diffusive constant coincides with the one obtained for the classical limit of D_q [8]: $D_{cl} \propto \varepsilon^2$.

The interpretation of the experimental results obtained in the presence of a monochromatic microwave signal is less straightforward. Strictly speaking, dynamical localization theory predicts that the ionization thresholds are time independent $(D_q = 0)$. In contrast with this theoretical conclusion our experimental results (Table I) show that the ionization thresholds depend on the duration of the microwave pulse. The time dependence once more obeys a power law but the experimental values of the exponent α lie in the interval between 0.13 and 0.27, quite far from the value $\alpha = 0.5$ which is typical for classical systems or for quantum systems in the presence of stochastic excitation. A first possible explanation of this experimental behavior may be the inadequacy of a onedimensional model used to picture the ionization process. Using the analogy of dynamical localization to Anderson localization [5] in solid-state systems it is known that localization of electronic wave functions in disordered solids is restricted to the one-dimensional case. For higher-dimensional systems it is possible to find extended states [12]. A second possible explanation is that the unavoidable presence of a small thermal noise destroys the full dynamical localization of the system [13]. In fact, for a physical process similar to our experiment it has been found that the diffusion coefficient of a quantum



FIG. 3. Ionization threshold fields $\varepsilon_{10\%}$ in laboratory units as a function of the initial principal quantum number n_0 for coherent signal excitation $\varepsilon_{c,10\%}$ and for broadband noise excitation $\varepsilon_{N,10\%}$. Interaction time $\tau = 5 \ \mu s$.

system subjected to an intense coherent field plus a small additive noise is proportional to $D_q \propto \varepsilon_N^2 \varepsilon_c^4$ [8]. Starting from this equation it follows that one has to expect for the $\varepsilon_{c,10\%}$ threshold fields a $\tau^{-\alpha}$ dependence characterized by the exponent $\alpha = 0.25$. This theoretical value of the exponent is roughly close to the values observed experimentally—with the exception of the state with $n_0 = 85$ (see Table I). It seems this state shows a higher stability with respect to microwave ionization. Similar phenomena have been observed in microwave ionization experiments with hydrogen atoms and have been interpreted as a manifestation of scars [14]. A third reason may be that coupling to the continuum may induce a slow decay of the Floquet states populated by the microwave [15].

Up to this point we focused on the interpretation of the experimental time dependence of the ionization process in the stochastic and in the monochromatic case. However, our study also shows another important experimental result: A broadband stochastic field covering the range of typical transition frequencies between neighboring atomic states is more effective than a coherent microwave field in the ionization of Rydberg atoms for equal values of the control parameters (initial principal quantum number. microwave power, and interaction time). This conclusion is manifested in Fig. 2 $(n_0 = 85)$ for all the investigated time intervals. To obtain further information about this effect, an estimate of the $\varepsilon_{N,10\%}$ threshold was made for several different states, with the interaction time fixed to 5 μ s, in the range $n_0 = 60-90$. The results of this investigation are plotted in Fig. 3. In the same figure we show for comparison the threshold fields $\varepsilon_{c,10\%}$ obtained with coherent excitation of frequency equal to 8867 MHz and with the same interaction time. In the investigated n_0 range, and for the chosen interaction time, the microwave power necessary to ionize 10% of the atoms is always

lower in the case of broadband noise than in the case of coherent signal excitation. This experimental observation shows that in a multiphoton ionization process, a pure broadband noise within the appropriate spectral range is more effective than a coherent signal. However, the question immediately arises which part of the spectrum of our broadband noise source is responsible for the observed ionization (in the absence of a coherent field) or for its enhancement (in the presence of a coherent field). It is therefore open to further experiments either to reduce the bandwidth of the noise or to select discrete frequencies [16] or small frequency bands from the large spectral range accessible, and to study the resulting ionization efficiency. This should provide further information on the necessary properties of perturbations used to destroy localization and to enhance or even to cause ionization.

In conclusion, our experimental study shows that quantum coherent phenomena are effective in rubidium Rydberg atoms excited by monochromatic microwave fields. We also show that a weak additive noise destroys these quantum properties after a certain interaction time. In addition, our study of the time dependence proves experimentally for the first time that a quantum system subjected to broadband excitation displays a diffusive behavior. We also give experimental evidence that broadband microwave excitation can more efficiently ionize Rydberg atoms than a coherent microwave field.

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