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Microwave Excitation of Rydberg Atoms in the Presence of Noise

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We study theoretically and experimentally the effect of noise on Rydberg atoms passing through a strong coherent microwave field. We derive a master equation containing the coherent field exactly. Its solution reveals the presence of four universal dynamical regimes: (i) an initial classical regime, (ii) a subsequent coherent localized regime, (iii) a transition, induced by the noise, in which coherence and localization are destroyed, and (iv) the final regime, where equidistribution over the quasienergy states is reached.

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Periodically driven nonlinear dynamical systems may behave chaotically, in a classical description, giving rise to diffusion of an appropriately chosen action variable.[1] A simple model system is provided by the standard map[2]; a physical example is a Rydberg atom in a strong, coherent microwave field.[3-7] In a quantum description, these systems mimic the classical dynamics only over a short initial time interval, after which quantum-mechanical interference effects change the dynamics,[8] leading in some cases to localization with respect to the action variable.[7,9,10] For the kicked rotor, this localization effect may occur for weak coupling as a perturbative effect,[11] or, for strong coupling, it may be interpreted as a form of Anderson localization.[9] In both cases it crucially depends on the coherence of the wave function.

The presence of a small stochastic contribution to the kicking force of the rotor was shown to be sufficient to destroy localization and to lead back to diffusion on long time scales.[12] Recently the effects of dissipation and fluctuations on the localization of the angular momentum of the kicked rotor have been investigated.[13,14] It was shown that the coherence of the initial state and the accompanying localization are destroyed on a time scale inversely proportional to the dissipation rate or noise intensity. For longer times it was found that the system is governed by noise-driven diffusion.

The main purpose of the present paper is to show both theoretically and experimentally that the main trends derived from the study of the kicked rotor also hold for the intensively studied case of Rydberg atoms in strong microwave fields. Additionally, the present paper represents the first experimental study of the influence of noise on coherent quantum effects. So far, the influence of a stochastic component could only be demonstrated through the change of the ionization probability of Rydberg atoms.[15] The particular setup of the experiment in Ref. 15 only allows one to study a fixed exposure time of the atoms to the microwave field. Therefore the transition between the regimes listed above was not observable. In the present Letter we present a complete theory and report on experiments which allow one to study the response of the Rydberg atom for variable exposure times. The experimental findings are in accord with the theoretical predictions.

We consider Rydberg atoms traversing a waveguide which is excited by a superposition of a coherent signal in its TE_{01} mode and a noisy signal. We assume that the Rydberg atoms, before they enter the waveguide in the x direction (i.e., parallel to the electric microwave field), have been prepared in a parabolic substate in the x

direction such that a 1D approximation to the atomic dynamics is permissible.[5,6] The Hamiltonian then takes the form

$$H = H_0 + H_{\rm int} + H_R , \tag{1}$$

with

$$H_0 = \frac{p^2}{2m} - \frac{e^2}{4\pi\epsilon_0}\frac{1}{x} + exF\sin(\omega t), \quad x > 0 ,$$

$$H_{\rm int} = \hbar \sum_i g(\omega_i) x (b_i + b_i^\dagger) , \tag{2}$$

$$H_R = \hbar \sum_i \omega_i (b_i^\dagger b_i + \tfrac{1}{2}) .$$

Here H_0 describes the 1D model of a hydrogen atom coupled to the electric microwave field with amplitude F and frequency ω. The coupling of the atom to the quantized modes of the waveguide is described by $H_{\rm int}$. The coupling of the waveguide modes to the atom is given by the strength function $g(\omega_i)$. In the limit of infinite length of the waveguide, the sum over i is replaced by an integral over the density of modes of the waveguide. The field of the free waveguide is described by the Hamiltonian H_R. The operators $b_i^\dagger$, b_i are the usual boson creation and annihilation operators. The population of waveguide modes is determined by the mechanism that generates the noisy component of the field. In the present formalism we can treat any given noise distribution, such as, e.g., electronic shot noise which is amplified and fed to the waveguide power supply,[15] or thermally induced blackbody radiation in the waveguide. For the theoretical discussion we consider the latter source, and take the temperature T as a measure for the ratio between the coherent and the stochastic components of the driving field.

In order to proceed with the Hamiltonian (1), we introduce the interaction picture with respect to $H_{\rm int}$, treating $H_0 + H_R$ as the unperturbed Hamiltonian. In the following we use the Floquet basis $|\alpha\rangle$ which exactly diagonalizes the atomic one-cycle propagator $U_0(\tau,0)|\alpha\rangle$:

$$U_0(\tau,0)|\alpha\rangle = e^{-i\mu_\alpha\tau}|\alpha\rangle . \tag{3}$$

$U_0(t',t)$ is the unitary time-evolution operator from t to t', generated by H_0, and $\tau = 2\pi/\omega$ is the microwave period. The μ_α are the quasienergies, which are chosen to lie in the interval $0 \le \mu_\alpha < \omega$. $H_{\rm int}$ is now treated perturbatively by the application of the Born-Markoff approximation in a standard way[16] to obtain a master equation for the reduced statistical operator of the hydrogen atom in the coherent microwave field. We emphasize that in the present approach the microwave field is treated exactly. Details of the derivation will be presented elsewhere.

In the interaction picture and basis, the master equation for the statistical operator $\tilde\rho$ reads

$$\langle\alpha|\dot{\tilde\rho}|\alpha\rangle = \sum_\beta (M_{\beta\alpha}\langle\beta|\tilde\rho|\beta\rangle - M_{\alpha\beta}\langle\alpha|\tilde\rho|\alpha\rangle) , \tag{4a}$$

$$\langle\alpha|\dot{\tilde\rho}|\beta\rangle = -\frac{1}{2}\sum_\nu (M_{\alpha\nu} + M_{\beta\nu})\langle\alpha|\tilde\rho|\beta\rangle, \quad \alpha \ne \beta . \tag{4b}$$

The rate coefficients $M_{\alpha\beta}$ in (4) depend on the quasienergy differences $\Omega_{\alpha\beta k} = |\Delta_{\alpha\beta k}|$, $\Delta_{\alpha\beta k} = \mu_\alpha - \mu_\beta + k\omega$, the mode density $\bar\rho(\omega)$ and the coupling function $g(\omega)$ of the waveguide, and the Fourier components of the dipole matrix elements

$$x_{\alpha\beta k} = \frac{\omega}{2\pi}\int_0^{2\pi/\omega} e^{-i\Delta_{\alpha\beta k}t}\langle\alpha|U_0^\dagger(t,0)xU_0(t,0)|\beta\rangle dt . \tag{5}$$

Explicitly,

$$M_{\alpha\beta} = \sum_{k=-\infty}^{\infty} [\gamma_{\alpha\beta k} + (\gamma_{\alpha\beta k} + \gamma_{\beta\alpha k}) n_{\alpha\beta k}] , \tag{6}$$

with

$$\gamma_{\alpha\beta k} = 2\pi\bar\rho(\Omega_{\alpha\beta k}) g^2(\Omega_{\alpha\beta k}) |x_{\alpha\beta k}|^2 \Theta(\Delta_{\alpha\beta k}) ,$$
$$n_{\alpha\beta k} = [\exp(\hbar\Omega_{\alpha\beta k}/k_B T) - 1]^{-1} , \tag{7}$$

where Θ is the step function and k_B is the Boltzmann constant. The solution of (4) is given by single decaying exponentials for the coherences $\langle\alpha|\tilde\rho|\beta\rangle$; the occupation probabilities $\langle\alpha|\tilde\rho|\alpha\rangle$ of the quasienergy states, on the other hand, relax to a steady state as a superposition of exponentials. Returning to the Schrödinger representation, the statistical operator ρ at integral multiples $N\tau$ of the microwave period is given by

$$\langle\alpha|\rho(N\tau)|\beta\rangle = e^{-i(\mu_\alpha - \mu_\beta)N\tau}\langle\alpha|\tilde\rho(N\tau)\beta\rangle . \tag{8}$$

In order to obtain some physical insight, we have solved (3)–(6) numerically for an ideal waveguide of rectangular cross section of side lengths $a = 1.9$ cm, $b = 0.95$ cm, and temperatures $T = 4$ and 300 K, coherently excited in the TE_{01} mode with amplitudes $F = 4$ and 8 V/cm and frequency $\omega = 2\pi \times 10.6$ GHz. The initial state was chosen as the eigenstate of the 1D hydrogen model with principal quantum number $n_0 = 71$. Twelve atomic bound states, ranging from $n = 69$ to 80, were taken into account as a basis for the expansion of the Floquet states. This way, the final results could be expressed in the basis of bound states of the hydrogen model. In Fig. 1 we plot the width function[10]

$$W(N,T) = \exp\left[-\sum_n P_n(N,T) \ln P_n(N,T) \right] ,$$

where $P_n(N,T)$ is the probability to find the principal quantum number state $|n\rangle$ in ρ at time $N\tau$ and temperature T. The width function W is a convenient measure for the number of hydrogen bound states contained in the ensemble ρ. Since initially only a single state is occupied, we have $W(0,T) = 1$. Figure 1 actually consists of 100 snapshots of the width function W at discrete

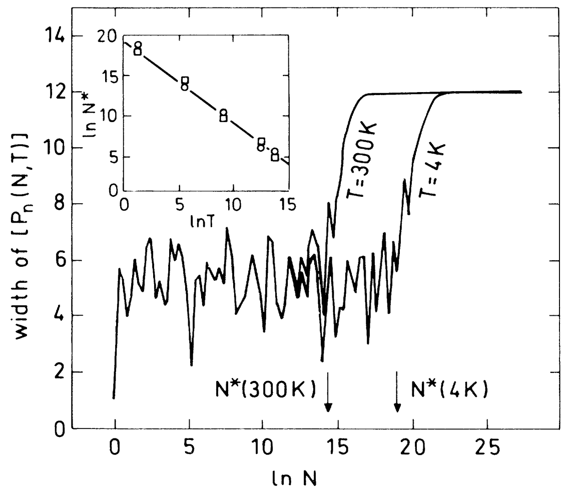

FIG. 1. Width of the $\{P_n\}$ distribution as a function of time for two different tempertures and $F=8$ V/cm. Inset: The relation between temperature and break time N^* for two different field strengths (squares, $F=4$ V/cm; circles, $F=8$ V/cm). The straight line interpolating the data was drawn to guide the eye, and reflects the relation $N^*T=$const.

times $N_j\tau$, $N_j=[10^{0.12j}]$, $j=1,\ldots,100$, the square brackets denoting the integer part. Four regimes of the dynamics can be clearly distinguished. In the first regime, occurring for very short times, the width W rapidly increases from 1 to a value between 5 and 6 (not resolved in Fig. 1). During this initial stage, quantum effects due to the discreteness of the quasienergies are not yet relevant. In the second phase, W saturates and oscillates quasiperiodically around some finite average. The fact that W is bounded by a number which is significantly smaller than the total basis size is the signature of localization. A transition which is sharp on the logarithmic scale of Fig. 1 follows the localization regime, in which nearly simultaneously the oscillations disappear and W increases abruptly, implying that coherence and localization are destroyed together at the transition time $N^*(T)$. A comparison of the two curves for $T=4$ and 300 K in Fig. 1 shows that the oscillations in the localized regime of the dynamics exactly reproduce each other, i.e., they are independent of temperature (noise power), but that the critical time N^* increases for decreasing temperature. In fact, the simple relation $N^*T=$const is obeyed by our numerical results over a vast range of temperatures, which is shown in the inset of Fig. 1. For $T\gg300$ K, the "temperature" merely serves as a convenient parametrization of the noise power as was explained above. The relation $N^*T=$const indeed follows from (6) and (7) in the limit $n_{\alpha\beta k}\sim k_BT/\hbar\Omega_{\alpha\beta k}\gg1$, where $M_{\alpha\beta k}\sim T$ holds. This limit is realized in our numerical examples. The fourth and final dynamical regime in Fig. 1 is the monotonic increase of W until equidistribution over the whole basis at $W=12$ is attained. This increase is much slower than the initial classical spreading of W, and it is due to noise-induced population transfer between mutually incoherent quasienergy states. This process is described by the master equation (4a) for the diagonal elements of the density matrix. Accordingly, the time scale in this regime must again scale inversely proportional to T, i.e., $W(N,T)=\tilde{W}(NT)$. This scaling property is clearly seen in Fig. 1, where the final rise of W follows nearly the same curve for different temperatures if the abscissa is shifted by the respective values of $\ln T$.

Summarizing our theoretical results, we have derived and solved a master equation for Rydberg atoms interacting with a strong microwave field in a noisy waveguide. Our predictions are that noise-induced destruction of coherence and of localization onsets at a critical interaction time N^*. It is followed by the redistribution of populations of quasienergy states which approaches equipartition after a very long time. This noise-induced diffusion is characterized by a time scale, which, like N^*, is inversely proportional to temperature or noise intensity. In the following we shall show some experimental evidence which supports these predictions.

In the present experiment, the atoms in a thermal beam of Rb Rydberg atoms sequentially pass through three spatially separated interaction regions. In the first region the atoms are laser excited to the 84 $P_{3/2}$ state. In the second one they are exposed to electronically shaped microwave pulses whose duration t can be continuously varied from ~10 ns to several tens of microseconds. Electronic shot noise (obtained from an idling traveling-wave tube amplifier[15] and ranging from 8 to 18 GHz) can be admixed to the coherent microwave pulses (carrier frequency 9.654 GHz) in arbitrary ratios. In the third region the atoms are ionized by an electric field ramp[17] (1.83 V/cm$\cdot\mu$s) and the electrons detected by use of a Channeltron multiplier. The timing of laser excitation, microwave interaction, and field ionization is performed in such a way that the atoms—independently of their particular velocity (Maxwellian velocity distribution)—interact for the same time duration with the microwave field. In the field-ionization region, the different Rydberg states are ionized at different times within the field ramp. Therefore $P(E;t)$—the ionization probability at ramp field E of atoms having interacted a time t with the microwave field—will be a unique function of the final n-state distribution of the Rydberg atoms. If we denote by $P^{(c)}(E;t)$ the response function of atoms which have interacted with the coherent microwave alone, and by $P^{(s)}(E;t)$ the response to a superposition of coherent signal and shot noise, then

$$\chi^2(t)\sim\int\left[\frac{P^{(s)}(E;t)-P^{(c)}(E;t)}{[P^{(s)}(E;t)]^{1/2}}\right]^2 dE$$

is a good measure for the effect of the noise. Figure 2 shows the $\chi^2(t)$ function for five different noise powers

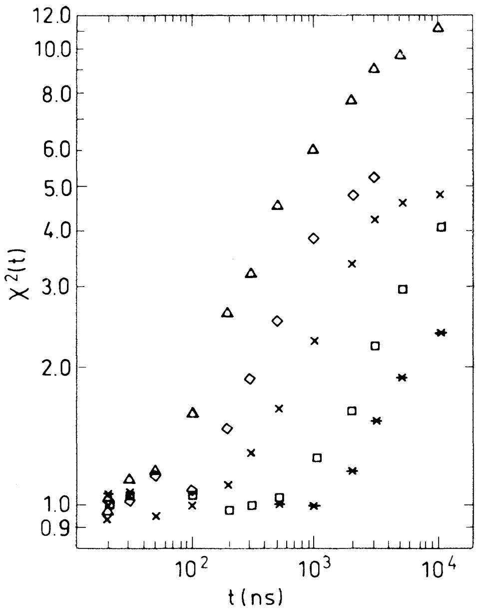

FIG. 2. χ^2 deviation (in arbitrary units) of the field-ionization response functions $P^{(c)}(E;t)$ and $P^{(s)}(E,t)$ of Rb atoms exposed to pulses of coherent and noisy microwaves as a function of the exposure time and several different noise powers. Triangles, 10 μW; diamonds, 4 μW; crosses, 1.6 μW; squares, 0.63 μW; stars, 0.25 μW. Signal power (3.2 mW) and noise powers were measured before entering the waveguide.

as a function of the exposure time t. The fast (classical) diffusive broadening of the initially prepared pure Rydberg state [regime (i)] occurs on a scale of a few field cycles ($t \ll 10$ ns) and is not shown in Fig. 2. Classical diffusion stops abruptly and results in the localization plateau [regime (ii)] which is followed by a linear rise of $\chi^2(t)$ [regime (iii)] where the noise takes over and a diffusive behavior prevails. For very long exposure time, $\chi^2(t)$ saturates, which corresponds to equidistribution of probability over the atomic states [regime (iv)]. The critical times t^* can be unambiguously extracted from Fig. 2. They are inversely proportional to the noise power (exponent, -1.07) which is in excellent agreement with the theoretical predictions. Moreover, for $t > t^*$, the slopes of the χ^2 curves in regime (iii) are nearly parallel which is consistent with the claim that the time scale of redistribution of population in the diffusive regime is inversely proportional to the noise power. The theoretical results can be trusted over an interaction time $\Delta t < \hbar/H_{\text{int}} \approx 1/gx$, where g and x are typical coupling constants and dipole matrix elements, respectively. In the present case $1/gx \approx 1$ ms and comparison to the theoretical results is allowed. As a result, the existence, as well as the proper scaling of a transition from the localization regime (short exposure time) to the stochastic diffusion regime (long exposure time), is quantitatively demonstrated.

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