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Manifestation of Localization in Noise-Induced Ionization and Dissociation.

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Abstract. – The manifestation of quantal localization for classically chaotic systems in disintegration processes such as ionization and dissociation is investigated. It is assumed that these processes are induced by noise. The effects of a nonperturbative mechanism that was recently introduced by D. Cohen are explored. The relation between these effects and noise-induced ionization of Rydberg atoms in the presence of microwave driving is discussed.

For several model systems [1-5] it was found that classical diffusion is suppressed by quantal effects that are similar to Anderson localization in disordered systems [6]. Some of the manifestations of these effects were observed in recent experiments [7-10]. In this letter the manifestation of quantum localization in the disintegration (for example ionization or dissociation) of systems due to external noise is studied. Understanding of such mechanisms is of great importance, since most experiments require the destruction of coherence for the purpose of observation. For the sake of concreteness the results will be presented first for the kicked rotor and finally the relevance for driven H and Rb atoms will be discussed [5, 7-13].

The standard model for investigation of the chaotic behaviour in systems with time-dependent Hamiltonians is the kicked rotor [1, 2, 6]. It is defined by the Hamiltonian

$$\mathscr{H} = \frac{1}{2}\hat{p}^2 + K\cos\theta \sum_m \delta(t-m), \tag{1}$$

where p is the angular momentum and its conjugate angle is θ that is confined to the interval $[0, 2\pi]$. The units are such that the moment of inertia and the time between the kicks are unity. The classical system is chaotic and exhibits diffusion in phase space [14] with the diffusion coefficient $D_{cl}(K)$ for $K > K_c = 0.9716$. For large values of K it is approximately

$$D_{cl} \approx \frac{1}{2}K^2. \tag{2}$$

When the rotor is quantized it exhibits Anderson localization in phase space [6]. In the quantum system the angular momentum takes the values $p_n = n\hbar$. The quasi-energy states are localized on the lattice of momentum states in the same way as energy states are localized on a one-dimensional disordered lattice [15]. The localization length for small $\hbar$ is [16]

$$\xi = \frac{D_{\mathrm{cl}}(K)}{2\hbar^2}. \tag{3}$$

This expression for the dynamical localization length holds for values of K that are larger than K_c but not too close to it. With small changes it applies also when $\hbar$ is not very small [16]. On scales smaller than ξ in momentum space diffusion with the classical diffusion coefficient D_{cl} takes place. On larger scales diffusion is suppressed and the system is localized [16]

The behaviour of the kicked rotor changes completely when noise is added [17-20]. The most interesting behaviour is found when the noise is weak. In this case the basic structure of localized states is preserved and the noise induces transitions between the localized states. The time t^* that is required to establish localization satisfies [16]

$$(\hbar\xi)^2 = D_{\mathrm{cl}}\, t^*. \tag{4}$$

If t_c is the time it takes to destroy coherence, then the noise is weak if

$$t^* \ll t_c. \tag{5}$$

If the noise is white, it induces a random walk on a lattice of localized states with spacing $\hbar\xi$ and hopping time t_c. It leads to diffusion with the diffusion coefficient [17, 20]

$$D_q \approx \frac{(\hbar\xi)^2}{t_c}. \tag{6}$$

This result was obtained from a careful derivation [17, 20]. It actually does not require that the noise is white, but merely that some correlations of the noise and the system are decoupled [20]. It was shown that it holds also for many forms of coloured noise that are of physical interest. From (6) and (4) one obtains a general expression for the diffusion rate,

$$D_q = \frac{t^*}{t_c} D_{\mathrm{cl}}, \tag{7}$$

that is much smaller than the classical diffusion coefficient.

Assume that the rotor disintegrates when it reaches momentum $p = p_{\max}$. This will occur if the rotor is a linear molecule, for example, and for $p > p_{\max}$ it dissociates. If initially the rotor is at $p = 0$, the time it takes for it to disintegrate in the presence of noise is of the order of

$$\tilde{t} = \frac{p_{\max}^2}{D_q}. \tag{8}$$

It is assumed here that $p_{\max} \gg \hbar\xi$ so that the disintegration is suppressed in the absence of noise. If the variance of the noise is ν, the coherence is destroyed when there is an appreciable probability for noise-induced transitions between levels. Consequently, $t_c = \hbar^2/\nu$ [17] and the resulting disintegration time satisfies

$$\tilde{t}^{-1} = \frac{t^*}{t_c}\tilde{t}_{cl}^{-1} \approx \xi^2 \frac{\nu}{p_{max}^2}, \tag{9}$$

where $\tilde{t}_{cl} = p_{max}^2/D_{cl}$ is the time of classical disintegration. Therefore (6), (2) and (3) imply

$$\tilde{t}^{-1} \approx C_1 K^4 \nu, \tag{10}$$

where $C_1 \approx 1/\hbar^4 p_{max}^2$ is a constant. If the disintegration is due to pure classical diffusion, one finds

$$\tilde{t}^{-1} \approx \tilde{t}_{cl}^{-1} \approx C_2 K^2 \tag{11}$$

with $C_2 = 1/p_{max}^2$ that is very different from (10). This type of disintegration is expected when $p_{max} \leq \hbar\xi$ or if the noise is sufficiently strong to destroy the phase coherence and the localization, but not sufficiently strong to dominate the transport ($1 \ll t_c \ll t^*$). The relation (10) is a clear manifestation of localization in trasport. Although the derivation was carried out for the kicked rotor, for the sake of concreteness, it should hold for a wide class of systems, although some details may have to be altered.

A system that exhibits localization like the rotor is the kicked particle. It is described by the Hamiltonian (1), but the angle variable θ is replaced by a space variable x that varies from $-\infty$ to $+\infty$. The momentum is not quantized, but the transitions that are induced by the kicks couple only momenta that differ by integer multiples of $\hbar$. The matrix elements are identical to those of the kicked rotor, therefore localization with the same localization length is found. If initially the particle is prepared in a state p_0, it can reach states only within a ladder $p_0 + n\hbar$, that is determined by p_0, where n are integers. Within the ladder, quantum localization takes place, and the dynamics are similar to those of the kicked rotor. Noise, however, induces transitions between ladders belonging to different values of p_0. Unlike the case of the kicked rotor the effect of noise on the kicked particle is *nonperturbative* in its nature, and therefore it leads to a much more effective destruction of localization, as shown by Cohen [21]. Due to noise-induced diffusion the spreading in momentum is $\Delta p \approx \sqrt{\nu t}$. The resulting variation of the velocities of the particles leads to a spread in position

$$\delta x \approx \frac{2}{3}\sqrt{\nu}\, t^{3/2}. \tag{12}$$

For the description of a state with localization length ξ in momentum, details on the scale $1/\xi$ are important. Consequently, localization is destroyed when δx exceeds $1/\xi$ resulting in

$$t_c \approx \frac{(9/4)^{1/3}}{\xi^{2/3}\nu^{1/3}}. \tag{13}$$

The quantal diffusion coefficient (6) is much larger for the kicked particle than for the kicked rotor due to the $\nu^{1/3}$-dependence on the variance of the noise. This result was verified numerically [21] and it is different from the perturbative result (9). The disintegration time following from (6) is

$$\tilde{t}^{-1} \approx \left(\frac{4}{9}\right)^{1/3} \frac{\hbar^2}{p_{max}^2} \xi^{2+2/3} \nu^{1/3}. \tag{14}$$

Therefore the disintegration time for the kicked particle satisfies

$$\tilde{t}^{-1} = C_3 K^{4+4/3} \nu^{1/3}, \tag{15}$$

where $C_3 \approx 1/\hbar^{10/3} p_{\max}^2$ is a constant. The dependence of $\tilde{t}$ on K is not very different from that of the kicked rotor but the dependence on the noise variance ν differs sharply.

In the presence of noise, the behaviour of real systems is intermediate between the kicked rotor and the kicked particle. Their proximity to each one of these idealized systems is determined by the number of ladders of states, such that the dynamics are restricted to within each ladder. The noise induces transitions between these ladders. Moreover, locally in phase space, many systems can be approximated by the kicked rotor [3, 5]. The most studied example of this type is the hydrogen atom in a microwave field [5, 7-13]. Let ω be the frequency of the microwave field. The driving microwave field induces transitions between levels that are separated approximately by ω in energy (in atomic units that will be used in what follows). These are called sometimes the photon states [5, 13]. The quasi-energy eigenstates are localized in the basis of these states. These are ladders of energy states that are not connected by the dynamics if one starts from a particular eigenstate of the unperturbed hydrogen atom. Noise induces transitions between these ladders. The proximity of the behaviour to the one of the kicked rotor or the kicked particle is determined by the number of these ladders g. This number g is approximately equal to the number of unperturbed states in the one-photon energy interval, so that for the one-dimensional hydrogen atom $g = \omega n^3$ where atomic units were used. This number g increases with the level number and therefore the kicked-particle approach becomes more applicable for initially excited level n_0 with a large value of $\omega_0 = \omega n_0^3 = g$. The excitation in microwave field can be approximately described by the Kepler map [5] which can be reduced locally to the standard map with Hamiltonian (1). From this reduction (see ref. [5], eqs. (13)-(18)) one obtains

$$K = 49\omega_0^{1/3} \varepsilon_0, \qquad k = \frac{K}{\hbar} = \frac{2.6\varepsilon_0 n_0}{\omega_0^{5/3}}, \qquad \hbar = \frac{K}{k} = 6\pi\omega_0^2/n_0, \tag{16}$$

where $\varepsilon_0 = \varepsilon n_0^4$, while ε is the field strength and $\hbar$ is the *effective* Planck's constant for this problem. Noise will be characterized by a decay rate Γ of the probability at a given level n, assuming that noise generates transitions mainly to nearby levels. For example, for the case of the black-body radiation at temperature T, one gets [22] $\Gamma \approx 4T/(3c^3 n^2)$, where the temperature and velocity of light are measured in atomic units. For given Γ the value of ν is determined by the change of the effective momentum $(\Delta p)^2 = \hbar^2 (\Delta N)^2$ of the electron in the Kepler map after one orbital period. Since the variation in number of photons is $\Delta N = \Delta n/\omega n^3$, one finds

$$\nu = (\Delta p)^2 = \frac{2\pi \Gamma n^3 \hbar^2}{\omega_0^2}, \tag{17}$$

where it was taken into account that the orbital period is $2\pi n^3$ and $(\Delta n)^2 = 2\pi \Gamma n^3$. It results from the assumption that transitions take place mainly to nearest levels. From (2), (3), (13) and (16), one obtains an expression for the coherence time (in the number of microwave periods)

$$\tau_c = \omega_0 t_c \approx \frac{2\omega_0^{5/3}}{k^{4/3} (\Gamma n^3)^{1/3} \hbar^{2/3}}. \tag{18}$$

Even for small values of Γ this time is relatively short, which leads to a fast ionization with a rate that is inversely proportional to τ_c. However, expression (18) can be applied only if the time τ_c is bigger than the time $\tau_\gamma = \omega/2\pi\Gamma$ required for transition to the nearest level, leading to

$$\frac{K^2}{45\hbar\omega_0} < \Gamma n_0^3 . \tag{19}$$

If this condition is not satisfied, then noise acts as in the case of kicked rotor and coherence time is inversely proportional to ν. The time it takes to ionize a fixed fraction, say 10%, of the atoms, in the presence of noise is proportional to $\tilde{t}$. If (19) holds eq. (15) with K replaced by ε applies, otherwise (10) should be used.

It is possible that the main predictions of this paper that are eqs. (10) and (15) have already been observed in experiments on rubidium atoms by the Munich group [23]. In these experiments Rb atoms are driven by a microwave field $\varepsilon \cos \omega t$ for a time t_i. In the first set of experiments localization was observed and its destruction by external noise was studied [9, 10]. In the second set of experiments [23], that was done for a different frequency, the field that is required to ionize 10% of the atoms, ε_c, was measured as a function of t_i, that is the time that the atoms interact with the microwave field. The experiment is performed in liquid-nitrogen temperature and no controlled noise is added. It was found experimentally that

$$\varepsilon_c \sim 1/t_i^\alpha , \tag{20}$$

where $\alpha = 0.25$ or $\alpha \approx 0.20$ depending on the initial state, where the value 0.25 was found for the lower initial states. According to the argument presented in this paper $\varepsilon_c \sim K$ and $t_i \sim \tilde{t}$. Therefore t_i is related to ε_c by eq. (10) or (15) with K replaced by ε_c and $\tilde{t}$ by t_i, depending on the mechanism that applies. The values that are predicted by eqs. (10) and (15) are $\alpha = 0.25$ and $\alpha = 3/16 = 0.188$, respectively, in agreement with the experimental results. Due to the close values of α predicted by these equations, it is hard to determine experimentally which of the two mechanisms, that were presented in the paper, takes place. The regime of applicability of (15) is restricted by (19) as well as by (5) and may be quite narrow. Therefore careful experiments are required to find a regime where it applies. Measurement of ε_c or t_i as a function of the noise variance ν will enable to distinguish between these mechanisms due to the big difference between the exponent of ν in eqs. (10) and (15).

It is not completely clear what is a possible source of noise in these experiments. One obvious possibility is thermal noise. Using the usual estimate for Γ due to black-body radiation for liquid-nitrogen temperature, we get $\Gamma n^3 \simeq Tn/c^3 \approx 10^{-8}$ for $n \simeq 100$. This is extremely small. Transitions to various l and m states induced by black-body radiation are not very effective due to the small phase space for low frequencies. The existence of other low-frequency noise combined with black-body radiation may induce transitions in a more effective way. Another possibility is that this noise results from the core electrons. The microwave field couples mainly to the outer electron. Due to exchange correlations with the core electrons the energy levels of the external electron are shifted by a very small amount leading to a shift in the quasi-energies. A very long time is required to resolve this shift and on short time scales of the experiment it looks as level widening or noise. The core electrons may act, therefore, as a heat bath for the external electron. Detailed calculations are required in order to evaluate the importance of this mechanism. Another possibility is that the noise originates from the microwave generator. In such a case it should depend on ε and the prediction of theory for α may be somewhat different, for example if ν is proportional to ε^2, one expects $\alpha = 1/6 \approx 0.17$ that is close to the experimental results. Since there is no classical theory for driven Rb atoms, we cannot rule out the possibility of an ionization mechanism that is of classical origin, making the theory presented in this paper inapplicable for the explanations of this experiment.

In summary, it was demonstrated how quantal localization manifests itself in noise-induced disintegration mechanisms, such as ionization and dissociation. The destruction of localization by noise may be particularly effective if it leads to transitions to states that are not coupled otherwise by the dynamics. However, the regime of applicability of this mechanism is found to be quite restricted. The theoretical arguments that were presented may be related to recent experimental results on the ionization of Rb atoms.

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