Algebraic decay of the survival probability in chaotic helium

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We demonstrate that the classical helium atom exhibits algebraic decay of the survival probability in the autoionizing eZe configuration. Excitation towards asymptotic orbits of marginal stability is identified as the cause of the nonexponential decay. Since one electron travels along a bound Kepler trajectory of infinite excitation in these orbits, we argue that the classical decay law will prevail in the quantum dynamics of a wave packet launched along the eZe configuration.

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The temporal decay of a dynamical system's survival probability $P_s(t)$ within a given phase-space volume, and its relation to the chaotic or mixed regular-chaotic nature of the dynamics have been subject to active research for more than one decade [1–14]. Whereas an *exponential* decay of $P_s(t)$ (or of correlations in general) indicates purely hyperbolic (i.e., colloquially, chaotic) dynamics, an *algebraic* time dependence is generally found when islands of regular motion are embedded in the chaotic sea. Due to the hierarchical structure of phase space in the boundary layer that encloses such islands [3,4], trajectories may remain trapped in their close vicinity over an appreciable amount of time. This delays the probability flow to remote domains of phase space and induces algebraic rather than exponential decay from a finite volume around the island. Indeed, a power-law decay $P_s(t)\sim t^{-\alpha}$ of the survival probability (with decay exponents in the range $\alpha\simeq 0.25\ldots 3.5$) was found in a number of studies on mixed regular-chaotic classical systems [1–7], as well as in theoretical and experimental studies on their corresponding quantum counterparts [8–10]. This led to the conclusion that the observed decay properties allow to identify the chaotic or mixed regular-chaotic nature of phase space, and to extract quantitative information on its local topology near regular islands.

However, it is worthwhile to note that trapping a trajectory near regular islands does not represent the only mechanism to induce algebraic decay of a classical system. Also close to *marginally* stable orbits (i.e., orbits with eigenvalues of the monodromy matrix equal to unity), which do not sustain a regular region in phase space, chaotic transport is considerably reduced, which leads again to an algebraic time dependence of the decay of correlations (which was shown, e.g., for the stadium billiard [11–13]). The present contribution now explicitly demonstrates that this latter mechanism of power-law decay becomes particularly relevant in the context of complex atomic systems. On the basis of a numerical integration of the classical equations of motion, we show that the classical helium atom in the eZe configuration—which we consider a paradigmatic case of such systems—exhibits algebraic decay of the survival probability. This algebraic decay does not arise due to regular islands in phase space (the classical dynamics of our three-body Coulomb system is fully chaotic), but rather due to the presence of asymptotic orbits of marginal stability, which are characterized by an extreme (infinite) excitation of one of the electrons. Since electrons launched along these asymptotic two-electron orbits evolve with negligible correlation over time scales that are asymptotically long, we obtain a good estimation of the decay exponent α from well-known scaling properties of the two-body Coulomb problem. Also quantum wave packets in the real atom will exhibit the classically observed decay, since the classical scaling laws prevail in a quantum description.

Let us start from the classical Hamiltonian of the helium atom, in atomic units, given by

$$H=\frac{p_1^2}{2}+\frac{p_2^2}{2}-\frac{Z}{|r_1|}-\frac{Z}{|r_2|}+\frac{1}{|r_1-r_2|}, \tag{1}$$

where r_i and p_i represent the positions and momenta of the electrons, and $Z=2$ the nuclear charge. Initial conditions $p_i=p_{xi}\mathrm{e}_x$, $r_i=x_i\mathrm{e}_x$, with $x_1>0$, $x_2<0$ (with e_x the unit vector along the x axis) define the eZe configuration in which the electrons are located along the x axis, on opposite sides of the nucleus (due to the Coulomb singularity of the interaction, the electrons cannot pass the nucleus in a collinear arrangement). Extensive studies have shown that the classical dynamics of this configuration is fully chaotic [15–18]. All bound periodic orbits within the collinear configuration are unstable, i.e., small perturbations increase with time and eventually lead to (single) ionization, as a consequence of the interelectronic repulsion. The survival probability $P_s(t)$ of this system is therefore naturally given by the probability for the atom to remain bound, after a given time t. In our numerical integration of the classical equations of motion derived from Eq. (1), we consider an electron to ionize as soon as it exceeds a given distance R_0 from the nucleus, i.e.

$$|r_i|>R_0, \quad \text{for} \quad i=1 \quad \text{or} \quad 2, \tag{2}$$

and, simultaneously, has a kinetic energy that is larger than the effective binding potential created by the nucleus and the other electron, i.e.,

$$E_i\equiv\frac{p_i^2}{2}-\frac{Z}{|r_i|}+\frac{1}{|r_1-r_2|}>0. \tag{3}$$

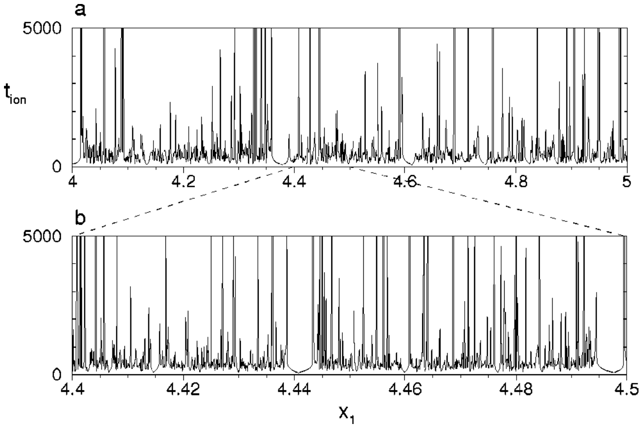

FIG. 1. Ionization time t_{ion} as a function of the initial position x_1 of one of the electrons. t_{ion} is plotted for 1000 equidistant values of x_1 within (a) $4 \leqslant x_1 \leqslant 5$ and (b) $4.4 \leqslant x_1 \leqslant 4.5$. The initial position of the other electron is adjusted such that the total energy equals $E = -1$; the initial momenta are zero. One clearly recognizes a fractal pattern of the ionization time as a function of the initial condition.

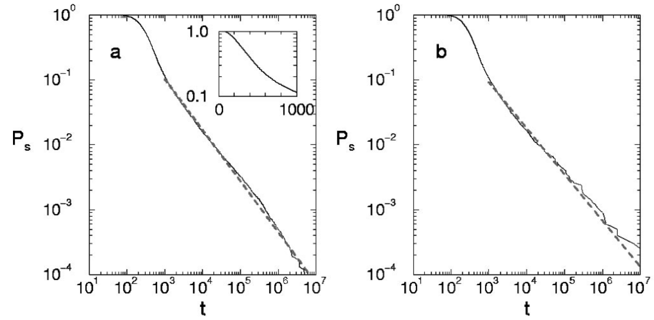

FIG. 2. Survival probability P_s of a classical two-electron atom in the eZe configuration, as a function of time. (a) Collinear configuration, with 100 000 equidistant initial values of $x_1 \in [4;5]$, and otherwise the same parameters as employed in Fig. 1. (b) Three-dimensional initial conditions, with 10 000 equidistant initial values of $x_1 \in [4;4.1]$, and $p_{y_1} = p_{z_2} = 0.1$, for a total energy $E = -1$. In both cases, P_s exhibits algebraic decay $P_s \sim t^{-\alpha}$ for $t \geqslant 1000$. $\alpha = 0.7868 \pm 0.0005$ (a), and $\alpha = 0.712 \pm 0.002$ (b) are extracted from the least-square fits indicated by the dashed lines in (a) and (b), respectively, in good agreement with the analytical estimate (8).

Provided R_0 is much larger than a typical distance of the order of $1/|E|$, characterizing bound orbits at a given total energy *E*, the electron *i* will never return to the nucleus if these two conditions are fulfilled (we chose $R_0 = 100$ in our calculations, at fixed energy $E = -1$).

In Fig. 1(a) we plot the ionization time as a function of the initial position x_1 of one of the electrons, which is varied in 1000 equidistant steps from $x_1 = 4$ to $x_1 = 5$ (the initial position x_2 of the other electron is adjusted such that the total energy equals $E = -1$; the initial momenta are zero). For each of these initial conditions, the equations of motion are propagated until the atom is ionized (a double Kustaanheimo-Stiefel transform is employed to regularize electron-nucleus collisions [16,19]). We clearly recognize a fractal structure of the ionization time as a function of the initial condition (see also Refs. [18,20]), which is further supported by the ''magnification'' in Fig. 1(b). The regular, cusp-shaped minima in the ionization time correspond to trajectories that come very close to triple collision events (where both electrons collide with the nucleus at the same time) [18]. Near those collisions, a huge amount of energy can be transferred between the electrons, which leads to rather fast ionization. On the other hand, Fig. 1 also exhibits divergent ionization times, and it is precisely those which dominate the asymptotic time dependence of the survival probability.

$P_s(t)$ is given by the fraction of trajectories that have not led to ionization after time *t*, as compared to the total number of trajectories starting from a given ensemble of initial conditions. In Fig. 2(a), this quantity is plotted for 100 000 equidistant initial positions x_1 within the range $4 \leqslant x_1 \leqslant 5$ (initial values of the other variables as in Fig. 1). We clearly see that P_s first decreases almost exponentially for $t \lesssim 500$, and then follows a power-law decay

$$P_s(t) \sim t^{-\alpha}, \tag{4}$$

for $t \gtrsim 1000$. A least-squares fit of P_s for times longer than $t = 1000$ yields the decay exponent $\alpha \simeq 0.79$. Similar decay curves (with very similar power-law exponents $\alpha \simeq 0.7 \ldots 0.85$) are not only found for different ranges of initial values x_1, but also for different values of the nuclear charge (as was verified for $Z = 1.5 \ldots 5$). Furthermore, the observed behavior does *not* arise as an artifact of the collinear arrangement of the configuration. Figure 2(b) shows the time-dependence of the survival probability for 10 000 trajectories that correspond to a true three-dimensional motion of the electrons. In contrast to Fig. 2(a), nonzero values $p_{y_1} = 0.1$ and $p_{z_2} = 0.1$ of the *transverse* momentum components along the unit vectors e_y and e_z, are chosen, for each value of x_1. Again we find an algebraic decay of the survival probability above $t = 1000$, with a decay exponent $\alpha \simeq 0.71$.

The physical cause of this delayed decay process can be identified by directly investigating trajectories with long survival times (Fig. 1), i.e., which contribute to the power-law tail of P_s (Fig. 2). A typical long-lived (collinear) trajectory is shown in Fig. 3. We see that the time-evolution of the atom is dominated by an extremely extended (but bound) Kepler trajectory to which one of the electrons is excited after several nearby collisions with the other electron. Only at the end of this Kepler orbit (at $t \simeq 44\,000$), the electrons continue to strongly interact with each other, which finally leads to ionization (at $t \simeq 45\,000$). Hence, for the major part of time, the classical system evolves in the vicinity of an *asymptotic* orbit, which is characterized by an infinite excitation of one of the electrons (i.e., one electron moves at an infinite distance from the nucleus with zero kinetic energy). As has been shown in Ref. [16], such asymptotic orbits exhibit *marginal* stability: The Lyapunov exponents of periodic orbits characterized by a highly excited Kepler trajectory of one of the electrons decrease to zero with increasing Kepler excitation, due to the asymptotic decorrelation of the electronic motion.

The decay exponent numerically obtained from Fig. 2(a) agrees quite well with this interpretation. Inspired by Fig. 3,

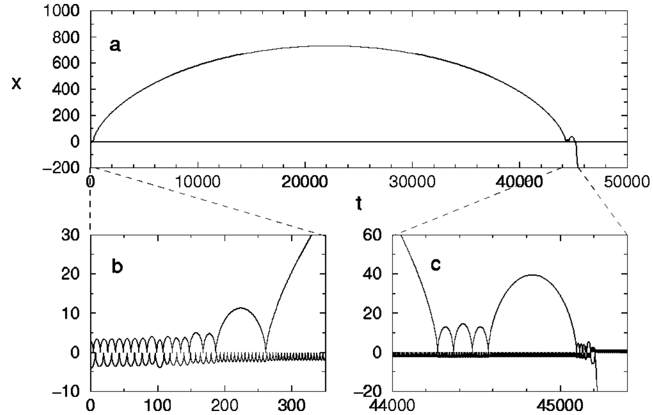

FIG. 3. A typical long-lived trajectory of the collinear eZe configuration. The positions of the electrons along the e_x axis are plotted as a function of time. Initial condition: $x_1=4.201\,07$, $p_{x1}=p_{x2}=0$, $E=-1$. We see that the time evolution of the atom is dominated by an extremely extended Kepler trajectory (a), to which one of the electrons is excited at $t\simeq 260$, after several nearby collisions with the other electron (b). At $t\simeq 44\,300$, the electron returns to the nucleus and collides with it several times, before the atom ultimately ionizes at $t\simeq 45\,200$ (c).

we approximate the survival time of the long-lived atom by the time

$$t=2\pi(Z-1)(-2E_i)^{-3/2}, \tag{5}$$

which either one of the electrons spends along a highly excited Kepler trajectory with effective energy E_i, Eq. (3). Assuming that, within a small interval below $E_i=0$, the probability $p(E_i)$ to excite an electron with the Kepler energy E_i is a smooth function of E_i (which is justified since the energy transferred at the collision does not sensitively depend on the critical amount necessary to eject one electron), we expand $p(E_i)$ in a Taylor series around $E_i=0$,

$$p(E_i)=p_0+p_1E_i+\frac{1}{2}p_2E_i^2+\cdots. \tag{6}$$

The probability for the atom to remain nonionized after time t then follows by integrating the contributions of all Kepler orbits with a period longer than t:

$$P_s(t)=\int_{-\tilde{t}^{-2/3}}^{0} p(E_i)dE_i \quad \text{with} \quad \tilde{t}=\frac{\sqrt{8}t}{2\pi(Z-1)}$$

$$=p_0\tilde{t}^{-2/3}-\frac{1}{2}p_1\tilde{t}^{-4/3}+\frac{1}{6}p_2\tilde{t}^{-6/3}-\cdots. \tag{7}$$

This leads to

$$P_s(t)\sim t^{-2/3}, \tag{8}$$

for long times t. Hence, few straightforward assumptions already reproduce an algebraic decay of the survival probability, with a decay exponent $\alpha=2/3$ fairly close to the one observed in the numerical calculation. Note that Eq. (8) neither accounts for the time that elapses before excitation to the highly excited Kepler orbit [see Fig. 3(b)], nor for the time between completion of the Kepler orbit and final ionization of the atom [see Fig. 3(c)]. These corrections to the survival time enhance the actual value of $P_s(t)$ with respect to the estimation (8), at given t. Since the relative weight of these corrections decreases with time, this leads to $\alpha\gtrsim 2/3$, in agreement with our numerical calculation.

A very similar decay phenomenon was observed in the classical s-wave helium model, where the electrons are restricted to spherical states [20]. The probability density of trajectories ionizing at time t was found to decay $\sim t^{-1.82}$ [20], which apparently corresponds to a power-law decay of the survival probability with a decay exponent $\alpha=0.82$, slightly above 2/3 as in our system. Asymptotic orbits analogous to the ones of Fig. 3 are responsible for this algebraic decay in s-wave helium. Within a statistical framework assuming purely chaotic dynamics, the decay exponent $\alpha=2/3$ was also derived for the Kepler map approximation of the driven hydrogen atom [14]. However, strongly driven one-electron atoms as considered in Ref. [14] exhibit regular phase-space domains embedded in the chaotic sea, which apparently induce a smaller decay exponent, $\alpha_H<2/3$, as suggested by numerical and laboratory experiments [9] ($\alpha_H\simeq 0.25\ldots 0.6$). Such regular domains do *not* exist in our present case of doubly excited states in the eZe configuration [22].

Let us finally address the question to which extent the observed decay law will manifest in the corresponding quantum system, i.e., in the doubly excited two-electron atom. We consider a minimum-uncertainty wave packet $|\psi_t\rangle$, initially launched in the regime of highly doubly excited states of helium. The quantum counterpart of the classical survival probability is then given by the bound probability P_b of this wave packet. Assuming [in analogy to the classical treatment (5)–(8)] that only highly excited states from Rydberg series below single ionization thresholds substantially contribute to P_b for long times, we have

$$P_b(t)=\sum_s\sum_{n=n_0}^{\infty}P_n^{(s)}\exp(-\Gamma_n^{(s)}t), \quad \text{for} \quad t\to\infty, \tag{9}$$

where n_0 is the principal quantum number of the outer electron above which the Rydberg series begins (i.e., above which the outer electron sees a modified hydrogen potential), $P_n^{(s)}=|\langle\psi_{t=0}|\psi_n^{(s)}\rangle|^2$ represents the initial overlap of the wave packet with the nth Rydberg state $|\psi_n^{(s)}\rangle$, s labels the associated ionization threshold, and $\Gamma_n^{(s)}$ is the autoionization width of $|\psi_n^{(s)}\rangle$. According to general scaling laws for Rydberg state matrix elements [21] (which are a direct consequence of the classical scaling laws of the two-body Coulomb problem), both $P_n^{(s)}$ and $\Gamma_n^{(s)}$ scale as n^{-3}, for large n. Hence, transforming the sum over n into an integral, Eq. (9) turns into

$$P_b(t)\sim\sum_s\int_{n_0}^{\infty}dn\, n^{-3}\exp(-\Gamma_0^{(s)}tn^{-3})$$

$$=\sum_s\frac{1}{3}(\Gamma_0^{(s)}t)^{-2/3}\int_0^{\Gamma_0^{(s)}tn_0^{-3}}dy\, y^{-1/3}e^{-y}, \tag{10}$$

and we again obtain an algebraic decay $P_b(t) \sim t^{-2/3}$ for long times $[t \gg (\Gamma_0^{(s)} n_0^{-3})^{-1}]$.

In conclusion, we predict an algebraic decay of the survival probability of doubly excited two-electron atoms in the *eZe* configuration. More generally, the described scenario is expected to apply for any highly excited Rydberg system, which—due to strong configurational coupling of its internal degrees of freedom or due to an external perturbation—lacks regular domains in the phase space of bound classical motion. Then only the excitation towards marginally stable Kepler orbits with asymptotically large excursions can prevent the atomic system from rapid disintegration, leading to an algebraic decay $\sim t^{-\alpha}$ with $\alpha \simeq 2/3$, which is obtained with the only ingredient of the scaling laws of the two-body Kepler problem. As a matter of fact, the recent experimental observation of population trapping in extremely highly excited Rydberg states of lithium under a short microwave pulse [23] provides a simple example of that mechanism: there, a strong microwave field occasionally promotes the Rydberg electron to highly excited orbits with recurrence times that are longer than the duration of the microwave pulse. Electrons transferred in such Kepler orbits are protected against ionization and should induce an algebraic decay $\sim t^{-2/3}$ as a function of the pulse duration, for field amplitudes that leave no atomic population in the domain of mixed regular-chaotic classical dynamics, i.e., in Rydberg states of moderate excitation. Since the quoted experiment can discriminate between the population of extremely high and moderate Rydberg levels, the predicted decay law is amenable to immediate experimental verification and can be compared to the decay dominated by mixed regular-chaotic dynamics [9,10].

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[1] C. F. F. Karney, Physica D **8**, 360 (1983).
[2] B. V. Chirikov and D. L. Shepelyansky, Physica D **13**, 395 (1984); Phys. Rev. Lett. **82**, 528 (1999).
[3] R. S. MacKay, J. D. Meiss, and I. C. Percival, Phys. Rev. Lett. **52**, 697 (1984).
[4] J. D. Meiss and E. Ott, Phys. Rev. Lett. **55**, 2741 (1985).
[5] C. F. Hillermeier, R. Blümel, and U. Smilansky, Phys. Rev. A **45**, 3486 (1992).
[6] Y.-C. Lai, C. Grebogi, R. Blümel, and M. Ding, Phys. Rev. A **45**, 8284 (1992); Y.-C. Lai, M. Ding, C. Grebogi, and R. Blümel **46**, 4661 (1992).
[7] G. Zumofen and J. Klafter, Phys. Rev. E **59**, 3756 (1999); S. Benkadda, S. Kassibrakis, R. White, and G. M. Zaslavsky, *ibid.* **59**, 3761 (1999).
[8] Y.-C. Lai, R. Blümel, E. Ott, and C. Grebogi, Phys. Rev. Lett. **68**, 3491 (1992).
[9] A. Buchleitner, D. Delande, J. Zakrzewski, R. N. Mantegna, M. Arndt, and H. Walther, Phys. Rev. Lett. **75**, 3818 (1995).
[10] G. Casati, G. Maspero, and D. L. Shepelyansky, Phys. Rev. Lett. **82**, 524 (1999); G. Benenti, G. Casati, G. Maspero, and D. L. Shepelyansky, *ibid.* **84**, 4088 (2000).
[11] F. Vivaldi, G. Casati, and I. Guarneri, Phys. Rev. Lett. **51**, 727 (1983).
[12] K.-C. Lee, Phys. Rev. Lett. **60**, 1991 (1988).
[13] H. Alt, H.-D. Gräf, H. L. Harney, R. Hofferbert, H. Lengeler, A. Richter, P. Schardt, and H. A. Weidemüller, Phys. Rev. E **53**, 2217 (1996).
[14] F. Borgonovi, I. Guarneri, and P. Sempio, Nuovo Cimento Soc. Ital. Fis., B **102**, 151 (1988).
[15] G. S. Ezra, K. Richter, G. Tanner, and D. Wintgen, J. Phys. B **24**, L413 (1991).
[16] K. Richter, G. Tanner, and D. Wintgen, Phys. Rev. A **48**, 4182 (1993).
[17] J.-M. Rost and D. Wintgen, Europhys. Lett. **35**, 19 (1996); J.-M. Rost, J. Phys. B **27**, 5923 (1994).
[18] X. Tang, Y. Gu, and J. Yuan, Phys. Rev. A **54**, 496 (1996); Z. Bai, Y. Gu, and J. Yuan, Physica D **118**, 17 (1998).
[19] S. J. Aarseth and K. Zare, Celest. Mech. **10**, 185 (1974).
[20] G. Handke, M. Draeger, and H. Friedrich, Physica A **197**, 113 (1993).
[21] Note that Ref. [10] predicts a decay exponent $\alpha_H = 1$ for mixed regular chaotic phase space structure. However, this result describes the leakage from the *local* vicinity of a nonlinear resonance island, through absorbing boundary conditions. In contrast, in the experiment [9] as well as in our present paper the survival probability is determined by the asymptotic transport to phase-space regions, which are *remote* from the initially occupied area.
[22] H. Friedrich, *Theoretical Atomic Physics*, 2nd ed. (Springer-Verlag, Berlin, 1998).
[23] M. W. Noel, W. M. Griffith, and T. F. Gallagher, Phys. Rev. Lett. **83**, 1747 (1999).