Multiple time scales for recurrences of Rydberg states

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An initially localized Rydberg wave packet, with $\Delta n \ll n$, spreads uniformly over its entire classical orbit within a time much larger than the classical period $T_{\rm cl}$. However, it eventually reassembles, with a hierarchy of recurrence times: $(n/3+\frac{1}{2})T_{\rm cl}$, $(n^2/4+\frac{1}{2})T_{\rm cl}$, etc. This phenomenon has no classical analog.

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Quantum systems with equidistant energy levels, such as harmonic oscillators, or particles precessing in a magnetic field, have a motion which is periodic in time. Surprisingly, excited hydrogen atoms also have a periodic behavior, in spite of the fact that their energy levels,

$$E_n = -me^4/2n^2\hbar^2 , \tag{1}$$

are not equidistant. This can be seen as follows.

Consider a hydrogen atom prepared in such a way that the positive-energy part of its spectrum is negligible (that is, ionized atoms are removed by the preparation procedure). We can then expand the state vector into bound-state eigenfunctions,

$$\psi(\mathbf{r},t)=\sum_n c_n u_n(\mathbf{r}) e^{-iE_n t/\hbar} , \tag{2}$$

where $\int u_n^* u_s d\mathbf{r}=\delta_{ns}$ and $\sum_n |c_n|^2=1$. Since this sum converges, a *finite* number of c_n can make $\sum_n |c_n|^2$ arbitrarily close to 1, and are therefore sufficient to represent ψ with arbitrary accuracy. As a consequence, any ψ is arbitrarily close to a periodic function of time [1]. In the present case, all the exponents in (2) have the form $2\pi it/T_0 n^2$ where $T_0=4\pi\hbar^3/me^4=3.04\times10^{-16}$ s. Let L be the least common multiple of all the n for which we do not neglect c_n. Obviously, ψ has a period T_0L^2.

This recurrence is *exact,* but the required time is enormous if many levels are appreciably excited. However, *nearly* exact recurrences occur considerably earlier. The recurrence probability is

$$P(t)=|\langle\psi(0)|\psi(t)\rangle|^2=\left|\sum_n w_n e^{-iE_n t/\hbar}\right|^2 , \tag{3}$$

where $w_n=|c_n|^2$. If the initial wave packet is well localized, its energy spread is small and the coefficients w_n are large only in a narrow domain of n (typically $\Delta n \sim \sqrt{n}$). Let N be an integer anywhere near the middle of that domain, and let $\nu=n-N$. We can expand

$$n^{-2}=N^{-2}-2N^{-3}\nu+3N^{-4}\nu^2-4N^{-5}\nu^3 +5N^{-6}\nu^4-\cdots . \tag{4}$$

If we keep only the first two terms, the exponents in Eq. (3) are $(2\pi it/T_0N^3)(2\nu-N)$. Apart from a common phase $-2\pi it/T_0N^2$, all these exponents become integral multiples of $2\pi i$ whenever

$$t=T_{\rm cl}=N^3T_0/2 . \tag{5}$$

This is the classical period of revolution for energy E_N, as expected. For short times, the quantum wave packet moves as a classical particle. For longer times, higher terms in (4) destroy the phase coherence, and the wave packet spreads over the entire orbit.

Yet, the wave packet eventually reassembles: if we take more than two terms in Eq. (4), the exponent in (3) becomes, apart from an irrelevant additive constant,

$$\exp[(\pi it/T_{\rm cl})(2\nu-3N^{-1}\nu^2+4N^{-2}\nu^3-5N^{-3}\nu^4+\cdots)] . \tag{6}$$

When $t=2NT_{\rm cl}$, the second term in this series yields an integral multiple of $2\pi i$, and the wave packet reappears at its original position. Actually, upon closer examination, it is found that this recurrence already occurs after $(N/3)+\frac{1}{2}$ classical periods (where N, which was only loosely defined above, has to be adjusted so as to be a

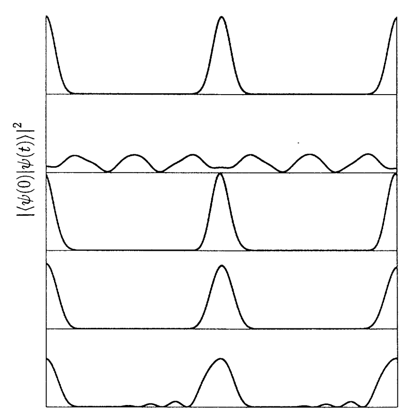

FIG. 1. Recurrences of a wave packet consisting of Rydberg states with $n=990$–1010. The value $|\langle\psi(0)|\psi(t)\rangle|^2$ is plotted vs time. The time at the center of each graph is, from top to bottom: $T_{\rm cl}$ (one classical period), $100T_{\rm cl}$ (a random time), $333.5T_{\rm cl}$ (first-order recurrence), $250\,000.5T_{\rm cl}$ (second-order recurrence), and $(2\times10^8+0.5)T_{\rm cl}$ (third-order recurrence). Each graph extends over two classical periods.

multiple of 3). Indeed, let $t=(N/3+\frac{1}{2})T_{\rm cl}$. The first two terms in (6) give

$$2\pi i\nu\left[\frac{N}{3}+\frac{1}{2}\right]\left[1-\frac{3\nu}{2N}\right]\simeq 2\pi i\nu\left[\frac{N}{3}-\frac{\nu}{2}+\frac{1}{2}\right]$$
$$=2\pi i\left[\frac{\nu N}{3}-\frac{\nu(\nu-1)}{2}\right]. \quad (7)$$

We can always adjust N so that $N/3$ is an integer. Also, $\nu(\nu-1)/2$ always is an integer. Therefore, apart from terms of order N^{-1}, the exponent in (6) is a multiple of $2\pi i$. The factor $N/3$ (without $\frac{1}{2}$) can also be obtained from semiclassical arguments [2]. This recurrence has been experimentally observed [3,4]. There also are "fractional revivals," namely partial recurrences, which occur for rational fractions (with small denominators) of the full recurrence time [5,6].

As time passes, the third term in the series in (6) gradually destroys these periodic recurrences, but new ones appear at integral multiples of $N^2T_{\rm cl}$. Here too, the same argument shows that the first such reappearance already occurs at $t=[(N^2/4)+\frac{1}{2}]T_{\rm cl}$, where N must again be adjusted to make it even, if necessary. These recurrences are then destroyed by the following term in (6), and reappear at integral multiples of $[(N^3/5)+\frac{1}{2}]T_{\rm cl}$, and so on. This hierarchy of recurrences is illustrated in Fig. 1 for the case $N=1000$, with 21 energy levels having a binomial distribution of weights (roughly a Gaussian distribution): $w_n=2^{-20}20!/(n-990)!(1010-n)!$.

An experiment indicating the existence of a third-order recurrence was performed by Parker and Stroud [7], who unfortunately overlooked the factor $\frac{1}{5}$ in $N^3T_{\rm cl}/5$. It is likely that sharper results could have been obtained if the waiting time after the laser pulse had been five times shorter.

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