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Two-Frequency Excitation of Hydrogen Atom

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Abstract—By using the Kepler map formulation of the microwave excitation of the hydrogen atom, we show that the introduction of a second incommensurate external frequency leads to a significant decrease of the delocalization border. Numerical computations confirm this prediction.

The problem of the excitation of an hydrogen atom under a monochromatic, linearly polarized, microwave field has attracted much interest in the past few years [1–10], in connection with the possibility of experimental investigation on the relevance of classical chaos in quantum mechanics. By now we have a sufficiently clear general picture of the quantum excitation process. In particular it was found [2, 4, 5] that the chaotic diffusion which takes place in the classical case and which leads to strong ionization can be suppressed by a localization phenomenon somewhat analogous to the one occurring in $1-d$ lattice problems of solid state physics. More precisely one can distinguish between two regimes defined by $\omega_0 < 1$ and $\omega_0 > 1$ (where $\omega_0 = \omega n_0^3$ is the ratio between the microwave frequency and the frequency $1/n_0^3$ of the unperturbed motion corresponding to the initially excited state n_0). For $\omega_0 < 1$ the excitation process is satisfactorily described by the classical dynamics. Instead, for $\omega_0 > 1$ quantum inhibitory effects produced by quantum interference are at work, and a stronger field than predicted by the classical chaos border is required for ionization [2].

This phenomenon of quantum suppression of classical chaos, which has been observed in recent laboratory experiments [11–13], turned out to be substantially stable for modifications of the model such as, e.g. the introduction of a second spatial dimension [3] or the introduction of an additional static field.

It is therefore interesting to investigate what would be the most efficient way to destroy this localization. A promising approach appears to be the introduction of a second perturbing frequency [14, 15] incommensurate with the first one. Indeed, the addition of such an incommensurate frequency was already shown to significantly reduce localization effects in simple models [16]. An heuristic explanation of this effect was provided by a formal identification of the quantum, deterministic time-dependent problem with a suitable lattice problem with a static pseudo-random potential [17, 18]. Indeed, under this identification the introduction of a second incommensurate frequency is equivalent to the introduction of a second spatial dimension in the lattice problem.

In this paper we present a theoretical and numerical study of the quantum excitation of a $1-d$ hydrogen atom in a microwave field with two frequencies. It is shown that also in this case the delocalization border is sharply reduced in the presence of a second

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incommensurate frequency, as is the 'local stability' at certain rational frequency ratios when $\omega_0 < 1$ [14, 15].

In order to analyze the dynamics of the $1-d$ hydrogen atom subject to two monochromatic fields with strength ε_1, ε_2 and frequency ω_1, ω_2, we used the Kepler-map formulation described in [5, 19]. This map gives the change of the phases $\phi_1 = \omega_1 t$, $\phi_2 = \omega_2 t \pmod{2\pi}$ of the field and of their conjugate variables N_1, N_2 over one orbital period of the electron, and is given by

$$\begin{aligned} \bar{N}_{1,2} &= N_{1,2} + k_{1,2} \sin \phi_{1,2} \\ \bar{\phi}_{1,2} &= \phi_{1,2} + 2\pi\omega_{1,2}[-2(\omega_1 N_1 + \omega_2 N_2 + E_0)]^{-3/2} \end{aligned} \tag{1}$$

where $k_{1,2} = 2.58(\varepsilon_{1,2}/\omega_{1,2}^{5/3})$ and $E_0 = (-1/2n_0^2)$ is the initial energy. In the quantum case $N_{1,2}$ would give the number of quanta absorbed from the fields (we use atomic units). In order to derive the four-dimensional map (1), it was assumed that $\omega_{1,2} n_0^3 > 1$ (see [5]). We will assume now that the two fields are of the same order of magnitude, so that the threshold for the onset of classical chaos in (1) will be at least not higher than the chaotic border for just one frequency:

$$(\varepsilon_{cr}) \sim \min \frac{1}{49\omega_{1,2}^{1/3} n_0^5}. \tag{2}$$

Therefore above the border (2), one has diffusive excitation in the classical model. If the ratio $\omega_2/\omega_1 = v \geq 1$ is integer, then $v\phi_1 - \phi_2 = \text{const}$ so that the phase ϕ_2 can be eliminated and the $4-$dim map reduces to a $2-$dim map in the variables ϕ_1 and $N = N_1 + vN_2$. The diffusion rate in N will be given by

$$D = \frac{(\Delta N)^2}{t} \sim \left(\frac{k_1^2}{2}\right)\left(1 + \frac{q^2}{v^{4/3}}\right); \quad q = \frac{\varepsilon_2}{\varepsilon_1} \tag{3}$$

where t is the number of mapping iterations.

In the quantum case, interference effects will produce exponential localization in the number of absorbed photons, with a localization length $l_\varphi \sim D$. We recall that this estimate is derived from a general prescription, that quantum diffusive behaviour up to a (discrete) time t is possible only if $\Delta N > t$, where ΔN is the number of excited states up to time t; in the present case, $\Delta N \sim (\Delta t)^{1/2}$ [5] which immediately yields $l_\varphi \sim D$ as the maximum spread in the number of absorbed photons. If this l_φ becomes comparable with the number of photons required for ionization: $N_I \sim 1/(2n_0^2\omega_1)$, then localization will be destroyed, and the excitation will be close to the classical one. From $l_\varphi \sim N_I$ we get the conditions for localization:

$$n_0 < \frac{\omega_0^{7/3}}{6.6\varepsilon_0^2(1 + q^2 v^{-4/3})} \tag{4}$$

where $\varepsilon_0 = \varepsilon_1 n_0^4$, $\omega_0 = \omega_1 n_0^3$. This result is almost the same as in the monochromatic case [4, 5], so that the introduction of the second field does not strongly change the localization picture.

As is well known, in the classical case, in the region of strongly developed chaos, the diffusion rate would practically not change in the case of incommensurate frequencies. However, in the quantum case the non-commensurability of frequencies produces a significant lowering of the delocalization border. In that case, indeed, the number of excited states $\Delta N_1 \Delta N_2$ grows according to $(k_1 k_2/2)t$. The delocalization condition is then $\Delta N_1 \Delta N_2 > t$. This condition requires $k_1 k_2 > 2$ to be compared with $k^2 \sim N_1$ for one

frequency case. It is therefore clear that with two incommensurate frequencies* a lower field is required for delocalization. Strictly speaking, even in that case one should not expect two-dimensional delocalization but rather localization over an exponentially large scale. Since, however, just a finite number of photons is needed for ionization, the condition $\Delta N_1 \Delta N_2 > t$ still defines the border for strong ionization. This condition can then be explicitly written:

$$n_0 > \frac{\omega_0^{5/3} v^{5/6}}{1.8 \varepsilon_0 q^{1/2}}. \tag{5}$$

In the range of n_0 given by (4) and (5) together, we expect a much stronger ionization with a two-frequency field than with a monochromatic one, or than with two commensurate frequencies.

Strictly speaking, the sharp difference between the commensurate and the non-commensurate case refers to an ideal situation with inifinite interaction time and with precisely specified frequency values. Quite clearly, in any physical situation, the finiteness of the interaction time will smooth this difference to some extent. In principle, it should be possible to numerically demonstrate the difference between commensurate and incommensurate cases, but the required computation time would be too long. We wish to recall in this connection that a distinction between commensurate and incommensurate cases occurs also with the well-known kicked rotator model. In spite of the simplicity of that model, a numerical investigation of the effects of commensurability required quite large computation times [21]. In the present model we expect the localization condition [4] to hold when the two frequencies are such that the system 'sees' a common period and the interaction time is long enough with respect to this common period. In practice, for interaction times of a few hundred cycles this will happen only when the frequency ratio is not too large an integer. Great care is needed in order to numerically check the above prediction (5). To this end, we made numerical experiments on the H-atom dynamics, by the same procedure described in [4]. The results are shown in Fig. 1. The full curve there shows the boundary of the parameter region defined by (5), with $q = 1$, $v = 1.382$. Here $2 - v$ is a rational approximation to the golden ratio and could be taken as a 'practically irrational' frequency ratio in our computations. Each point in Fig. 1 represents the outcome of a numerical simulation with different parameters in the range $66 \leq n_0 \leq 400$, $0.014 \leq \varepsilon_0 \leq 0.04$, $1.3 \leq \omega_0 \leq 2.5$ and over a time ~ 250 periods. The transition from localization to delocalization was detected by the increase of the ratio between quantum excitation probability $W_{1.5}^{q}$ above $\bar{n} = 1.5 n_0$ and the corresponding classical probability $W_{1.5}^{cl}$. The figure shows that this transition took place across the theoretical border (5) (full line).

Notice that the '1-frequency' border (dotted curve) is much higher. The three cases denoted by r in Fig. 1 correspond to a frequency ratio $v = 2(q = 1)$; in that case the crossover to strong ionization appears to take place at higher field values, about the threshold (4).

Some typical data are singled out in Table 1. In particular, we see that the case with two commensurate frequencies (third case) gives almost the same ionization as the one frequency case with the same total power.

*In any physical situation, the frequencies are not exactly known, so that the distinction between the commensurate and the incommensurate case is, strictly speaking, meaningless. Nevertheless, the dynamics of an atom interacting with the bichromatic field for a finite interaction time will follow the predictions of the incommensurate case if, in the given interaction time, it will not be able to perceive any periodicity in the perturbing field.

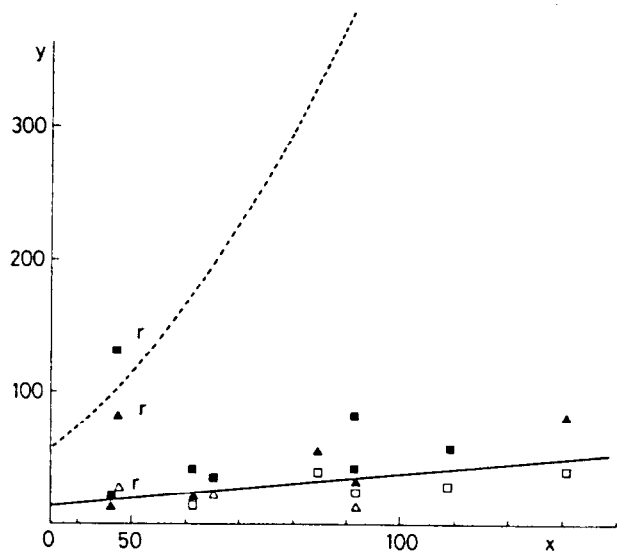

Fig. 1. The two frequency delocalization borders given by equation (4) (dotted curve) and equation (5) (full line) in the plane of the parameters $y = n_0/2\omega_0$, (number of photons required for ionization), $x = \omega_0^{2/3}/\varepsilon_0$. Here $q = 1$, $v = 1.382$. Full squares correspond to situations in which a value of $W = \log W_{1.5}^{q}/W_{1.5}^{cl}$ between -0.5 and 0. was obtained; full triangles, to $-1.0 \leq W < -0.5$; open squares to $-1.5 \leq W - 1$; open triangles to $W < -1.5$. The three cases with $x \approx 48(\varepsilon_0 = 0.021)$ and denoted by letter r, correspond to two commensurate frequencies with $\omega_0 = 1$ and $v = 2$, $q = 1$.

In this paper we illustrate how the localization effects that limit the diffusive classical-like excitation of a H-atom in a microwave field can be destroyed by the introduction of a second non-commensurate frequency.

Because experiments with single-frequency driving [11, 13] have shown for $\omega_0 > 1$ clear quantal suppression of classical chaos, supporting our earlier theoretical predictions [2] for this effect, and experiments with two-frequency driving have been reported for $\omega_0 < 1$ [14], it will be most interesting to have these two-frequency experiments extended into the $\omega_0 > 1$ region treated in this paper [22].

The considerations developed above for this particular problem are actually of a more general nature and can be used also for more general models to describe the excitation of systems with a level density ρ under the action of external fields with more than one frequency. Indeed for the monochromatic case, this situation was investigated in general in Refs. [5, 19] and it was shown that localization must be expected with a localization length in the number of absorbed photons given by

Table 1. Ionization probability $W_{1.5}$ after $\tau = 240$ microwave periods for the case $n_0 = 200$. The monochromatic case in the last line corresponds to the same total power as in lines 2, 3 and to the same peak intensity as in line 1

$\varepsilon_1 n_0^4$	$\omega_1 n_0^3$	$\omega_2 n_0^3$	$v = \frac{\omega_2}{\omega_1}$	$q = \frac{\varepsilon_2}{\varepsilon_1}$	$W_{1.5}$
0.014	2.5	3.382	1.3528	1	$1.5 \cdot 10^{-4}$
0.02	2.5	3.3825	1.3528	1	$3.0 \cdot 10^{-2}$
0.02	2.5	5	2	1	$1.5 \cdot 10^{-3}$
0.028	2.5	—	—	—	$2.5 \cdot 10^{-3}$

$$I_\varphi \sim 2\pi^2\mu^2\varepsilon^2\rho^2 \tag{6}$$

where μ is the one-photon matrix element and ε is the field strength. It was also shown that expression (6) gives essentially a diffusion coefficient in the number of absorbed photons. Instead, for two non-commensurate frequencies, the number of absorbed photons of both frequencies grows in time according to $\Delta N_1 \Delta N_2 \sim \pi \mu_1\varepsilon_1\mu_2\varepsilon_2\rho\, t$. According to our views [18], introduction of new non-commensurate frequencies is equivalent to the introduction of new degrees of freedom. Then, the above situation corresponds to a $2-d$ localization problem in solid-state physics, where the localization length, for $(D_1D_2)^{1/2} > 1$, grows exponentially with $(D_1D_2)^{1/2}$, where $D_{1,2}$ are diffusion coefficients for each frequency given by (6) [17]. On account of this analogy, we expect $\ln I_\varphi \sim (D_1D_2)^{1/2} = 2\pi^2\mu_1\mu_2\varepsilon_1\varepsilon_2\rho^2$.

Along the same lines, we are led to predict that the introduction of a third incommensurate frequency would produce a transition from localized to diffusive behaviour for $2\pi\mu\varepsilon\rho > 1$, where we assumed the three fields to be of the same order. This transition would be analogous to the Anderson transition in $3-d$ solid state problems [18]. According to this theoretical argument based on similarity with solid state physics localization problems, the introduction of a third incommensurate frequency should be even more effective for ionization. In this case, the threshold for strong ionization should be the same as the quantum border of stability [20].

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REFERENCES

1. J. E. Bayfield and P. M. Koch, *Phys. Rev. Lett.* **33**, 258 (1974).
2. G. Casati, B. V. Chirikov and D. L. Shepelyansky, *Phys. Rev. Lett.* **53**, 2525 (1984).
3. G. Casati, B. V. Chirikov, I. Guarneri and D. L. Shepelyansky, *Phys. Rev. Lett.* **59**, 2927 (1987).
4. G. Casati, B. V. Chirikov, I. Guarneri and D. L. Shepelyansky, *Physics Report*, **154**, 77 (1987) and references therein.
5. G. Casati, I. Guarneri and D. L. Shepelyansky, *IEEE J. Quant. Electron. Quant. Nonlinear Optics Single Atoms, Ions, Electrons* **24**, 1420 (1988).
6. J. E. Bayfield and L. A. Pinnaduwage, *Phys. Rev. Lett.* **54**, 313 (1985).
7. P. M. Koch, Electronics and atomic collisions, in *Proceedings of the XV ICPEAC, Brighton, England (1987)*, edited by H. B. Gilbody *et al.*, p. 501. Elsevier, Amsterdam (1988).
8. R. Blümel and V. Smilansky, in *The Structure of Small Molecules and Ions*, edited by R. Naaman and Z. Vager. Plenum Press (1988).
9. R. S. Mackay and J. D. Meiss, *Phys. Rev.* **A37** 4702 (1988).
10. R. V. Jensen, J. C. Leopold and D. Richards, *J. Phys.* **B21**, 1527 (1988).
11. J. E. Bayfield and D. W. Sokol, *Phys. Rev. Lett.* **61** 2007 (1988).
12. J. E. Bayfield, G. Casati, I. Guarneri and D. V. Sokol, *Phys. Rev. Lett.* **63**, 364 (1989).
13. E. J. Galvey, B. E. Sauer, L. Moorman, P. M. Koch and D. Richards, *Phys. Rev. Lett.* **61**, 2011 (1988).
14. L. Moorman, E.J. Galvez, B. E. Sauer, A. Movtazawi-M, K. A. H. van Leeuwen, G. V. Oppen and P. M. Koch, *Phys. Rev. Lett.* **61** 771 (1988).
15. R. Blümel, G. Jaeckel and V. Smilansky, Ionization of high n H-atoms by bichromatic microwave fields, Preprint.
16. D. L. Shepelyansky, *Physica* **D8**, 208 (1983); T. Geisel, *Phys. Rev.* **A41** 2989 (1990).
17. D. L. Shepelyansky, *Physica* **D28** 103 (1987).
18. G. Casati, I. Guarneri and D. L. Shepelyansky, *Phys. Rev. Lett.* **62**, 345 (1989).
19. G. Casati, I. Guarneri and D. L. Shepelyansky, *Phys. Rev.* **A36** 3501 (1987).
20. E. V. Shurak, *Zh. Eksp. Theor. Fiz.* **71**, 2039 (1976).
21. G. Casati, J. Ford, F. Vivaldi and I. Guarneri, *Phys. Rev.* **A34** 1413 (1986).
22. A Buchleitner, L. Sirko and H. Walther, Dynamical localization in the microwave interaction of Rydberg atoms and the influence of noise and bichromatic fields, Preprint.