RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 39, NUMBER 1 JANUARY 1, 1989

Ionization of high-n H atoms by bichromatic microwave fields

R. Blümel, G. Jaeckel, and U. Smilansky

Max-Planck-Institute for Quantum Optics, 8046 Garching, Federal Republic of Germany and Department of Nuclear Physics, The Weizmann Institute of Science, 76100 Rehovot, Israel

(Received 8 August 1988)

We present the first quantum-mechanical calculations of the ionization probability of high-n H Rydberg atoms perturbed by strong *bichromatic* microwave fields. The quantum-mechanical ionization thresholds agree very well with the experimental findings. The effect of rational approximants to the experimental frequency ratio is studied. We provide evidence to show that for the range of principal quantum numbers and field parameters investigated experimentally, the response of the H atoms to the bichromatic microwave field is quasiperiodic.

The ionization of high-n H atoms by microwave fields is relevant to central issues in two seemingly unrelated fields of physics: it is a study case for the "quantum chaos" community, and it is the most transparent example for the study of multiphoton processes in free atoms. Because of the relative simplicity of this problem, and because of the availability of accurate and comprehensive experimental data,[1-6] this subject attracted much attention[7-17] and received the status of a paradigm in both these fields.

Experiments investigating the response of hydrogen Rydberg atoms to strong bichromatic microwave fields are being conducted now and their results have been reported only recently.[18-20] From the theoretical point of view, this system bears on a fundamental issue concerning the nature of the response of a quantum system to a quasiperiodic perturbation. When a second frequency is switched on, the system ceases to be invariant under discrete time translations and hence, Floquet's theorem or any generalization thereof does not necessarily hold. There is no general theory by which one could analyze problems with quasiperiodic driving. Recent studies of some particular examples[21-24] reveal that the response may show a transition from quasiperiodic to aperiodic behavior, and an adaptation of these results to other problems of interest is far from being straightforward.

In the present Communication we present for the first time results obtained from the classical and quantal theories of the response of a realistic system to a quasiperiodic driving force. Both theoretical approaches will be compared with the experimental findings. We shall show that the physical insight and the numerical methods developed for the single-mode case[11] can be extended to the bichromatic case. The theoretical results reproduce the experimental data very well.

In a typical experiment,[1-6,18-20] a beam of neutral H atoms is excited to a specific high-n state and collimated to a microwave cavity, where it is exposed to a time-dependent linearly polarized electric field, which is characterized by its frequency (or two frequencies) and corresponding peak amplitude(s). Frequencies and field strengths are denoted by ω_i and ε_i, respectively ($i=1,2$). The passage time of the atoms through the cavity is typically of the order of a few hundred field cycles.

One measures the ionization probability $P_I(\varepsilon_1,\varepsilon_2,\omega_1,\omega_2;n_0)$ keeping the frequencies fixed and varying the field strengths for a range of initial quantum numbers n_0. At low fields there is no ionization ($P_I\approx 0$), while for high fields one achieves complete ionization ($P_I\approx 1$). The transition between these extremes occurs rather abruptly, and the onset of ionization is characterized by the field values for which P_I reaches the value of 10% for the first time. In a small number of cases, where P_I is not a monotonic function, this assignment might be ambiguous.

Our quantum-mechanical calculations are performed within the framework of the one-dimensional (1D) model[14] of H Rydberg atoms governed by the Hamiltonian

$$H(z,t)=H_0+z[\varepsilon_1\sin(\omega_1 t)+\varepsilon_2\sin(\omega_2 t)]\,, \tag{1}$$

$$H_0=\begin{cases}\dfrac{p^2}{2}-\dfrac{1}{z}, & z>0,\\ \infty, & z\le 0.\end{cases} \tag{2}$$

Here p and z are the momentum and position of the atomic electron in the field direction.

To introduce the notations and some essential concepts, we shall start by discussing briefly the theory of monochromatic excitation. One then deals with the Hamiltonian (1) where either $\varepsilon_1=0$ or $\varepsilon_2=0$. The fact that the perturbation is *periodic* in this case is of tremendous help. By Floquet's theorem we know that at any moment

$$|\Psi(t)\rangle=\sum_\alpha e^{i\chi_\alpha t}|\phi_\alpha(t)\rangle\langle\phi_\alpha(0)|n_0\rangle\,, \tag{3}$$

where time is expressed in units of the field period and χ_α are the "quasienergies." $|\phi_\alpha(t)\rangle$ are periodic and we abbreviate the notation by denoting $|\phi_\alpha(0)\rangle$ by $|\alpha\rangle$. If the quasienergy (QE) spectrum is discrete, the wave function $|\Psi(t)\rangle$ evolves in a quasiperiodic way. The QE spectrum is always discrete if the Hilbert space in which the system evolves is of finite dimension. In more general situations this will happen if the QE states $|\alpha\rangle$ are normalizable (localized).

After N cycles of the field the transition amplitude is given by

$$\langle n|\Psi(N)\rangle=\sum_\alpha\langle n|\alpha\rangle e^{i\chi_\alpha N}\langle\alpha|n_0\rangle\,. \tag{4}$$

A transition to a high-n state and subsequent ionization is

possible if and only if there exists at least one $|\alpha\rangle$ state which overlaps appreciably with *both* the n_0 and the high-n state. This is why the localization properties of the atomic states $|n\rangle$ in the $|\alpha\rangle$ basis are of such importance.

To characterize the degree of localization of an atomic state $|n\rangle$ in the QE basis we introduce the width function[11,12]

$$W(n)=\exp\left[-\sum_{\alpha}|\langle n|\alpha\rangle|^2\ln|\langle n|\alpha\rangle|^2\right]. \quad (5)$$

For $\omega n^3<1$, $W(n)$ shows a typical threshold behavior as a function of n for given ε and ω [see, e.g., Fig. 1(a) in Ref. 12]. States below the threshold are too narrow in the $|\alpha\rangle$ basis [$W(n)\approx 1$] and they have no chance to ionize. For states located above the threshold, $W(n)$ is large and strong ionization is observed.

The full line in Fig. 1 displays the relation between the field strength ε and the corresponding n value at which the jump in $W(n)$ occurs. The dashed line represents experimental ionization thresholds for the same field parameters. The agreement between the two curves is striking. Therefore, determining the transition points in the width function turns out to be a simple and powerful tool for the calculation of ionization thresholds in the single-frequency case.[11,12]

Turning now to the bichromatic driving we propose to take the following line of action. Any given irrational frequency ratio can be approximated by rationals p_n/q_n such that $|\omega_2/\omega_1-p_n/q_n|<\text{const}/q_n^2$. Replacing the irrational frequency ratio by its rational approximation in (1), we get problems with periodic driving, which, by Floquet's theorem, possess well-defined quasienergies and eigenvalues. It seems reasonable to suggest that by increasing the order of the approximants one obtains in the limit the corresponding quantities for the case of the irrational frequency ratio. Moreover, if we study the system during a *finite* time, one can approximate the solution of the quasiperiodic problem to any desired accuracy by the solution of a periodic problem with sufficiently large q_n.

TABLE I. First four rational approximants to the experimental frequency ratio (Refs. 19 and 20) and their relative deviations (%).

ω_2/ω_1	Relative error (%)
$\frac{3}{2}=1.5$	4.35
$\frac{11}{7}=1.571\,43$	0.21
$\frac{58}{37}=1.567\,57$	0.038
$\frac{69}{44}=1.568\,18$	0.0014
Experiment $=1.568\,16$	. . .

In this spirit we analyzed one of the ionization experiments reported in Refs. 18–20 where $\omega_1=7.58175$ GHz and $\omega_2=11.8894$ GHz, $\varepsilon_2=58.7$ V/cm (peak field value on the cavity axis) and ε_1 was swept. In our calculations we used the first four rational approximants to the experimental frequency ratio shown in Table I. In the experiment the fields are homogeneous in z direction, but vary considerably (of the order of 10%) in r direction because of the modal structure of the fields in the cavity. This implies that the theoretical ionization probabilities have to be averaged over the experimental field distribution. We have done this for the sweep field ε_1 in the case of the rational approximants $\omega_2/\omega_1=\frac{3}{2}$ and $\frac{11}{7}$. A simultaneous average over ε_2 is prohibitively expensive. Therefore, we chose the reduced value $\varepsilon_2=52$ V/cm as a representative of the average. The quantum-mechanical calculations are performed in a basis of 150 bound states including the full coupling to the continuum as described in Ref. 11. The interaction time was 100 cycles of the high-frequency mode. In Fig. 2 we show a detailed comparison between classical, quantum mechanical, and experimental ionization probabilities for fixed ε_2 as a function of the sweep field ε_1. All four rational approximants give rather similar ionization probabilities. Note, however, that the lowest rational approximant ($\omega_2/\omega_1=\frac{3}{2}$) fails to produce the

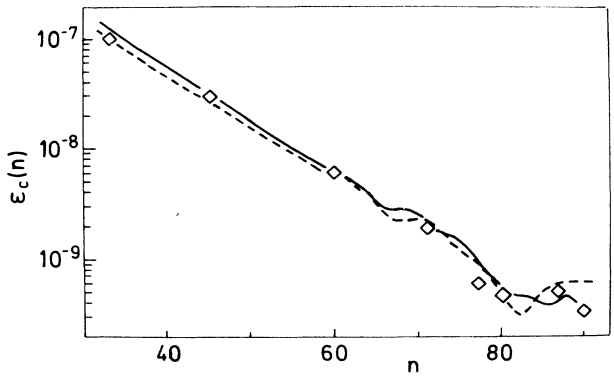

FIG. 1. Critical ionization fields as a function of principal quantum number n. Full line: critical fields extracted from numerically calculated width functions. Dashed line: experimentally determined fields (Refs. 1–6). In both cases $\omega=9.92$ GHz. Plot symbols: critical fields extracted from numerically calculated width functions for the bichromatic case. $\varepsilon_1/\varepsilon_2=2$, $\varepsilon_1+\varepsilon_2=\varepsilon_c$.

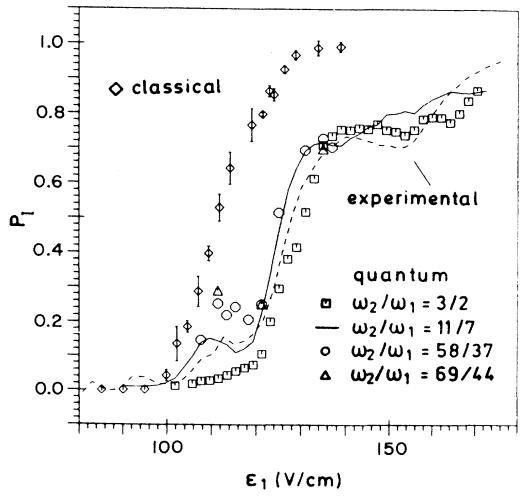

FIG. 2. Comparison between classical, quantum mechanical, and experimental ionization curves for bichromatic driving. The initial state is $n_0=43$. For the classical calculations $\omega_2/\omega_1=\frac{11}{7}$.

"bump" peaked at $\varepsilon_1 = 110$ V/cm. Away from this structure, where the calculations show a rather high sensitivity to field strengths and frequencies, the higher rational approximants reproduce each other to a good average accuracy. This observation was confirmed in calculations with a smaller basis which permits us to calculate full ionization curves even for the $\frac{58}{37}$ and $\frac{69}{44}$ approximants.

Table I shows that the fraction $\frac{11}{7}$ reproduces the experimental frequency ratio to 2.1 parts per thousand. Within a period of a few hundred cycles the system cannot resolve such a small frequency difference and we felt safe to perform the following quantum calculations using for the frequency ratio the value $\frac{11}{7}$ (compare also Fig. 2).

For $n_0 = 41, \ldots, 45$ and $\varepsilon_2 = 0$, 18.6, 40.7, 58.7, 92.5 V/cm we calculated the quantum-mechanical locus of (scaled) ionization threshold fields $\varepsilon_1 n_0^4$. Figure 3 shows that the quantum results are consistent with the experimental results and the classical theory—except that the classical thresholds are systematically too low. This behavior is analogous to the case of monochromatic driving[11] and probably due to the fact that for both frequencies used here, the classically scaled values satisfy $\omega_i n_0^3 < 1, i = 1,2$. This is the regime of parameters where the perturbation is slow on the atomic time scale. In adiabatic situations the maximum value obtained by the field is the relevant parameter, and here it is the sum of the two (scaled) fields. Indeed, the data in Fig. 3 seem to be correlated by $\varepsilon_1 n_0^4 + \varepsilon_2 n_0^4 = 0.12$ on the average. Fluctuations that are due to specific resonance conditions, resulting in nonmonotonic structures in the ionization curves, are definitely noticeable.

In the case of monochromatic driving, the existence of thresholds in the width function $W(n)$ was a useful shortcut for the calculation of ionization thresholds avoiding lengthy and expensive calculations which include the continuum. Detailed numerical experiments revealed that the width function of the "rationalized" bichromatically driven H atom also exhibits marked thresholds which are very similar to the ones encountered in the single frequency case. The locations of the two-frequency thresholds depend only on the sum $\varepsilon_1 + \varepsilon_2$ of the (scaled) field strengths of the two driving modes. Fixing the ratio $\varepsilon_1/\varepsilon_2 = 2$ and

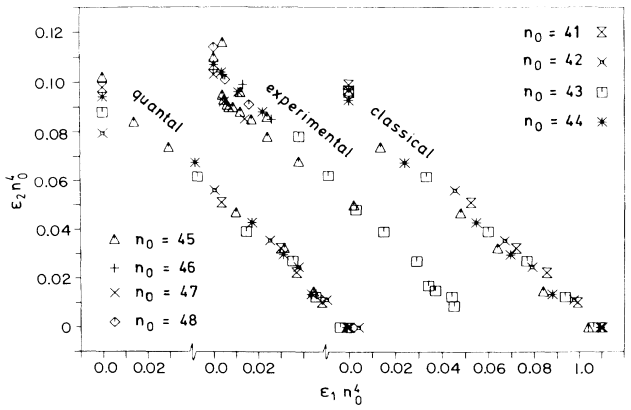

FIG. 3. The locus of the scaled 10% ionization threshold fields. From left to right: quantum mechanical, experimental (Refs. 18–20), and classical.

with $\omega_2/\omega_1 = \frac{11}{7}$, we determined the positions n of the thresholds of the width function for seven values of the (summed) field strength $\varepsilon = \varepsilon_1 + \varepsilon_2$. The result is displayed by the plot symbols in Fig. 1. The agreement with the single-frequency data is striking. This result, obtained for a special ratio of $\varepsilon_1/\varepsilon_2$, should be valid more generally. In order to test this conjecture, we calculated the sum of the field strengths of the two driving microwave modes for all n_0 values where experimental two-frequency ionization threshold data were available, and found it to be very close to the corresponding single-frequency critical fields $\varepsilon_c(n)$. This observation in connection with the numerical threshold results (see plot symbols in Fig. 1) leads us to the following prediction: Once experimental data for higher principle quantum numbers n_0 will be available (but still $\omega_1 n_0^3, \omega_2 n_0^3 < 1$), the sum field of the two microwave modes at threshold, plotted against the principle quantum number n, should collapse on the single-frequency data as displayed, e.g., in Fig. 1.

A prominent feature in the experimental data are the nonmonotonic structures ("bumps") in the ionization curves near threshold. The quantum calculations reproduce the positions and the relative heights of these structures. Within the statistical errors they are completely missing in the classical results. In the case of monochromatic driving, we have shown that the "bumps" are due to the clustering of a large number of avoided crossings in the quasienergy spectrum, a fact recently confirmed in Ref. 25. This is a purely quantal phenomenon and can be encountered only if the QE spectrum is discrete.[11,12] The appearance of bumps in the bichromatic case and our ability to reproduce them using a rather low rational approximant for ω_2/ω_1 is very significant. It indicates that in the present case, the generalized Floquet expansion[26,27] converges. In this case, the bumps are due to avoided crossings of the generalized Floquet eigenvalues, and the wave function is quasiperiodic in time.

In conclusion we may say that our understanding of the microwave ionization of H atoms in the domain $\omega n_0^3 < 1$ is rather complete. The study of bichromatic driving confirms the theoretical ideas and computational devices which were developed for the interpretation of the ionization induced by periodic fields.

The situation in the regime $\omega n_0^3 > 1$ may have some surprises in store. In this regime the theory for the periodically driven quantum problem predicts suppression of ionization due to localization of the QE states.[14] The localization phenomenon is sensitive to the quantum phases between matrix elements of the evolution operator, which may be drastically affected when a second field is switched on. This, and other related questions, are now under study.

We are grateful to Professor P. M. Koch for showing us his experimental data prior to publication and for stimulating discussions. This research was supported in part by a grant from the United States-Israel Binational Science Fund, Jerusalem, Israel, and by the Israeli Basic Research Commission.

[1]J. E. Bayfield and P. M. Koch, Phys. Rev. Lett. **33**, 258 (1974).
[2]J. E. Bayfield, L. D. Gardner, and P. M. Koch, Phys. Rev. Lett. **39**, 76 (1977).
[3]P. M. Koch, J. Phys. (Paris) Colloq. **43**, C2-187 (1982).
[4]P. M. Koch, in *Proceedings of the NATO Advanced Research Workshop on Fundamental Aspects of Quantum Theory, Como, Italy, 1985,* edited by V. Gorini and A. Frigerio (Plenum, London, 1986).
[5]K. A. H. van Leeuwen, G. v. Oppen, S. Renwick, J. B. Bowlin, P. M. Koch, R. V. Jensen, O. Rath, D. Richards, and J. G. Leopold, Phys. Rev. Lett. **55**, 2231 (1985).
[6]P. M. Koch, in *The Physics of Phase Space: Nonlinear Dynamics and Chaos, Geometric Quantization and Wigner Function,* edited by Y. S. Kim and W. W. Zachary, Lecture Notes in Physics, Vol. 278 (Springer-Verlag, Berlin, 1987).
[7]N. B. Delone, B. P. Krainov, and D. L. Shepelyansky, Usp. Fiz. Nauk. **140**, 355 (1983) [Sov. Phys. Usp. **26**, 551 (1983)], and references therein.
[8]J. G. Leopold and I. C. Percival, J. Phys. B **12**, 709 (1979).
[9]R. V. Jensen, Phys. Rev. Lett. **49**, 1365 (1982); Phys. Rev. A **30**, 386 (1984); in *Proceedings of the Workshop on Photons and Continuum States of Atoms and Molecules, Cortona, Italy, 1986,* edited by N. K. Rahman, C. Guidotti, and M. Allegrini, Springer Proceedings in Physics, Vol. 16 (Springer-Verlag, Berlin, 1987).
[10]J. G. Leopold and D. Richards, J. Phys. B **18**, 3369 (1985); **19**, 1125 (1986); O. Rath and D. Richards (unpublished).
[11]R. Blümel and U. Smilansky, Z. Phys. D **6**, 83 (1987).
[12]R. Blümel and U. Smilansky, Phys. Rev. Lett. **58**, 2531 (1987).
[13]R. Blümel and U. Smilansky, Phys. Scr. **35**, 15 (1987).
[14]G. Casati, B. V. Chirikov, D. L. Shepelyansky, and I. Guarneri, Phys. Rev. Lett. **56**, 2437 (1986); **57**, 823 (1986); Phys. Rep. **154**, 77 (1987).
[15]R. Blümel and U. Smilansky, in *Proceedings of the Conference on Chaos and Related Non-Linear Phenomena,* edited by I. Procaccia (Plenum, London, 1987).
[16]J. N. Bardsley, B. Sundaram, L. A. Pinnaduwage, and J. E. Bayfield, Phys. Rev. Lett. **56**, 1007 (1986); J. N. Bardsley and M. J. Comella, J. Phys. B **19**, L565 (1986).
[17]D. Richards, J. Phys. B **20**, 2171 (1987).
[18]L. Moorman, E. J. Galvez, B. E. Sauer, A. Mortazawi-M., K. A. H. van Leeuwen, G. v. Oppen, and P. M. Koch, Bull. Am. Phys. Soc. **32**, 1264 (1987).
[19]P. M. Koch, in *Proceedings of the Fifteenth International Conference on the Physics of Electronic and Atomic Collisions,* edited by H. B. Gilbody, W. R. Newell, F. H. Read, and A. C. H. Smith (North-Holland, Amsterdam, 1987).
[20]L. Moorman, E. J. Galvez, B. E. Sauer, A. Mortazawi-M., K. A. H. van Leeuwen, G. v. Oppen, and P. M. Koch, Phys. Rev. Lett. **61**, 771 (1988).
[21]D. L. Shepelyansky, Physica **8**, 208 (1983).
[22]M. Samuelides, R. Fleckinger, L. Touziller, and J. Bellisard, Europhys. Lett. **1**, 203 (1986).
[23]J. M. Luck, H. Orland, and U. Smilansky, J. Stat. Phys. (to be published).
[24]Y. Pomeau, B. Dorizzi, and B. Grammaticos, Phys. Rev. Lett. **56**, 581 (1986). See also R. Badii and P. F. Meier, *ibid.* **58**, 1045 (1987).
[25]H. P. Breuer, K. Dietz, and M. Holthaus, Z. Phys. D **8**, 349 (1988).
[26]T. S. Ho and S. I. Chu, J. Phys. B **17**, 2101 (1984).
[27]C. S. Chang and P. Stehle, Phys. Rev. A **4**, 641 (1971).