Localization of Classically Chaotic Diffusion for Hydrogen Atoms in Microwave Fields

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New experimental results are presented for short-pulse microwave ionization of highly excited hydrogen atoms. A comparison of these results with quantum numerical computations and analytical predictions provides for the first time experimentally grounded evidence of the localization phenomenon that leads to the suppression of the quantum version of the chaotic diffusion in action space occurring in the classical limit.

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The onset of chaotic motion in externally driven classical systems is a key problem in nonlinear dynamics that is now well understood in its essential aspects. In many physically interesting cases, as the perturbation strength increases beyond some critical value, the system starts absorbing energy in a diffusive way.[1] The question whether "diffusive" excitation processes can take place also in externally driven quantum systems is then a very interesting one for the physics of atoms and molecules in external electromagnetic fields.[2] This is a deep question involving the nature and the validity of the quasiclassical approximation when the underlying classical dynamics is chaotic.

The question of "quantum diffusion" was first addressed in a simple model system, the kicked rotator.[3] It was chosen because the features of classical chaos in it were relatively clean and well understood. Besides that, the numerical simulation of its quantum dynamics could be easily accomplished.[3,4] The major indication of this model[3,4] was that quantum mechanics suppresses the classical chaotic diffusion in action via a destructive interference effect similar to that responsible for the Anderson localization well known in condensed matter physics.[5] However, unlike Anderson localization, this new type of localization does not require the introduction of random elements from the outside; to stress this essential difference, it was called "dynamic localization." It was then submitted that dynamic localization should not be considered an artifact of the peculiar kicked rotor model, but a general effect that would eventually stop any diffusive quantum excitation, unless the related growth in time of the number of populated excited states is fast enough.[6,7]

The microwave ionization of the hydrogen atom is an ideal testing ground for this localization theory. Since the first experiments,[8] increasingly accurate numerical simulations of a classical model for electron motion under the combined influence of a Coulomb field plus a monochromatic oscillating electric field have shown that the experimentally observed thresholds for the onset of ionization follow the classical thresholds for the onset of chaotic instability, when the ratio ω_0 of the microwave frequency ω to the unperturbed Kepler frequency is less than 1.[9] Most theoretical analyses up to recent times were carried out on a simplified one-dimensional model, which is adequate for experimental situations in which the atom is initially prepared in a state very extended along the direction of the field.[10] Numerical simulations of the quantum dynamics of this model in the region $\omega_0 < 1$ confirmed the agreement between classical and quantum thresholds.[7,11] On the other hand, in the region $\omega_0 > 1$, localization theory applied to the one-dimensional model predicted that, due to the localization phenomenon, the quantum threshold for ionization should rise above the classical one, following a "quantum delocalization" border.[6,7,12] These predictions were supported by extensive numerical simulations of the one-dimensional quantum dynamics.[6,7,12,13]

A formulation of the classical dynamics at $\omega_0 \gtrsim 1$ by means of an appropriate map, the so-called "Kepler map,"[12] made it possible to find a connection between the one-dimensional hydrogen atom model and the kicked rotor model, and thus provided even firmer grounds for the application of localization theory. Moreover, by constructing a similar Kepler map for a two-dimensional model it was shown that the classical excitation in this model develops along very similar lines as in the one-dimensional model, due to the existence of an approximate integral of the motion which practically decouples the two degrees of freedom. This circumstance enforced the prediction that a localization theory very similar to the one-dimensional one should apply in the two-dimensional case also. Therefore, this localization theory predicts that a localization phenomenon should be experimentally observable even for atoms initially prepared in nonstrictly one-dimensional states and with magnetic quantum number (with respect to the direction of the field) $m=0$,[12] as is the case for the experiments described in this paper.

Presented here are new experimental results and the comparison of these results with numerical computations and analytical predictions. These reveal for the first time the localization phenomenon through an average nearly linear increase of the ionization threshold with increasing ω_0, with the rate of increase having the predicted value.

The use of fast atom beams for the study of microwave ionization of highly excited hydrogen atoms is a well established technique.[8,14-17] First, a nearly monoenergetic proton beam with particle kinetic energy typically near 23 keV is converted to a mixed-state neutral hydrogen atom beam by means of charge-transfer collisions. State-selective laser transitions in static electric fields are then used to select atoms in the beam with parabolic quantum numbers $n,n_1,m=7,0,0$ and excite them to a state with numbers $n_0,0,0$.

In the present experiments the static field strength was reduced to $F_S=0.87$ V/cm in the microwave field region, which would static-field ionize $n,0,0$ atoms only for n above 164. This field reduction was observed to produce a scrambling of hydrogen atom substates. The scrambling was due to electric field components before and after the microwave region that were not along the nominal electric field direction, vertical in the laboratory. As the apparatus had reflection symmetry through the vertical plane containing the atom beam axis, no scrambling in m should have occurred. A field-ionization scan of the substate-scrambled beam was precisely fitted with an almost uniform spread in n_1 between 0 and $\frac{3}{4}n_0$, taking $m=0$. This "two-dimensional" situation is to be compared with that of "three-dimensional" experiments that utilize almost statistically weighted scrambling in both n_1 and m.[15,17]

As the laser-excited atoms passed through holes in the sidewalls of a TE_{10}-mode microwave interaction waveguide (WR-62), they were exposed to a microwave pulse with a nominal envelope $\sin[\pi t/(7.5\text{ nsec})]$. The peak microwave field ranged from 2.5 to 3.8 V/cm at $n_0=98$ to 13 to 21 V/cm at $n_0=64$, depending upon ω, which was varied over the band 12.4 to 18 GHz. Machining the holes reduced the measured waveguide power transmission by only 4%. Hence leakage of microwaves out of the holes produced an additional long tail on the envelope of about 1% the peak field strength. Numerical calculations indicated that the added presence of this tail did not significantly change the predicted ionization probabilities.

The atomic beam leaving the microwave region was in a microwave-induced distribution of final atom quantum states that included the continuum. The beam then passed through a region of static field F_L which was adjustable and would ionize $n,0,0$ atoms with n above a corresponding adjustable value $\bar{n}$. It then entered a microwave ionization cavity field for converting all remaining excited atoms with n above 58 into protons again. The apparatus was designed to detect just these cavity-field-produced protons, and not protons produced, for instance, in the waveguide region. Therefore the ionization probability was defined experimentally as the reduction in cavity-field proton production rate induced by the waveguide microwaves, and hence was the sum of waveguide-induced probabilities for transitions to final bound states above $\bar{n}$ and for final continuum states. The initial-state quantum number n_0 was varied between 64 and 98. The value of $\bar{n}$ was increased with n_0 so as to keep their ratio roughly equal to 1.5.

A search was carried out for any alteration of ionization probabilities arising from residual in-band microwave noise, with negative results. A tunable narrow-band filter for $\omega=16$–18 GHz reduced the rms noise field strength from 4% to 0.06% and possibly increased sine-wave field strength values for 10% ionization probability by only (3 ± 3)%. Another check of the apparatus found that preionization of the excited atoms in a static field before the waveguide region reduced the proton detector signal down to the same background level to within 2% for the waveguide microwaves either on or off. In addition, the observation that $F_S=0.87$ V/cm gave microwave ionization probabilities similar to those at still lower values of F_S was taken to mean that the static electric field in the wave guide was not playing a major role in the results now to be reported.

In Fig. 1 we present thresholds for 10% ionization obtained experimentally and by quantum numerical simulations. These thresholds are defined as the field strengths at which a 10% probability was observed above the cutoff values $\bar{n}$. The error bars shown on a few of the experimental data points are the final rms uncertainties for averages over several runs taken on different days. A further ±10% uncertainty in the overall microwave field

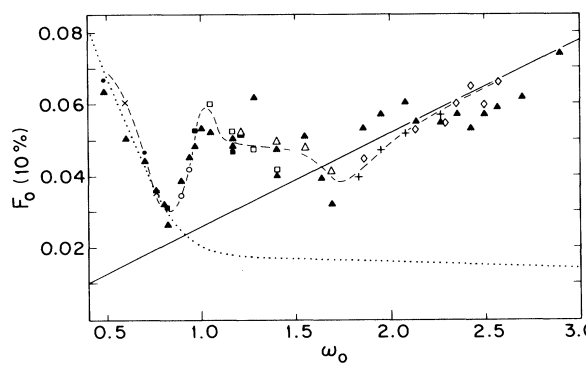

FIG. 1. A comparison at identical parameter values of experimental and quantum-mechanical values for the microwave field strength for 10% ionization probability, as a function of microwave frequency. The field and frequency are classically scaled, $\omega_0=n_0^3\omega$ and $F_0=n_0^4F$. Ionization includes excitation to states with n above $\bar{n}$. The theoretical points are shown as solid triangles. The dashed curve is one drawn through the entire experimental data set shown in Fig. 2. Values of n_0, $\bar{n}$ are ●, 64, 114; ×, 68, 114; ○, 71, 114; ■, 76, 114; □, 80, 120; △, 86, 130; +, 94, 130; ◇, 98, 130. Multiple theoretical values at the same ω_0 are for different compensating experimental choices of n_0 and ω. The dotted curve is the classical chaos border. The solid line is the quantum 10% threshold according to localization theory for the present experimental conditions.

strength is not included in the error bars. The numerical calculations were made in one dimension, but otherwise closely simulated the important experimental conditions and choices of parameters, including F_S and the pulse envelope. Two theoretical borders are also plotted in the figure. One of them (dotted curve) is the chaos border. (In the region $\omega_0 < 1$, the chaos border is extrapolated smoothly from the region $\omega_0 \approx 1$ to lower values of ω_0.) The other border (solid line) is the quantum theoretical prediction obtained for the present experimental situation by following the analytical procedure described in Ref. 13. This border is the field strength for a 10% probability flow into levels above the cutoff value, before the setting up of the final localized distribution. This quantum border is obtained by inserting the analytical expression for the localization length into the quantum equation for the exponential final-state distribution, integrating the latter above the cutoff value, and requiring the result to be a 10% probability. The result for the present experimental conditions is $F_0(10\%) = (0.23\omega^{1/6})\omega_0$, which exhibits only a weak $\omega^{1/6}$ deviation from classical scaling that amounts to about 8% over the present experimental range in ω.

The overall agreement between experimental and quantum numerical calculations seen in Fig. 1 is quite good, the overall rms deviation between these being 12%. For $\omega_0 > 1.5$ both sets of data rise with increasing ω_0, independent of particular values chosen for n_0 and ω. This almost linear rise is seen to agree in slope and location with the prediction of the analytical theory for localization. It also exhibits the predicted near-classical scaling. To be emphasized is that the analytical theory does not describe either quantum or classical resonance effects, as it is based upon only the smoothed behavior of stationary state distributions in quantum number or classical action. Also one should not expect the weak signs of narrow structure seen in the quantum numerical results to coincide with experiment, as in a static field; two-dimensional atom distributions have a different and more smooth averaged quantum energy-level structure than do one-dimensional atoms. Yet the high-frequency agreement between experiment, quantum calculations, and the analytical theory of localization is quite remarkable.

The experimental and quantum numerical results in the low-frequency region $\omega_0 \leq 0.8$ are consistent with theory, which predicts no localization for these frequencies. This region and the intermediate frequency region $0.8 \leq \omega_0 \leq 1.5$ are best discussed by including a comparison with classical numerical computations; see Fig. 2. For $\omega_0 \leq 0.8$, classical and quantum numerical values closely agree, and on average are 14% and 8% below experiment, respectively. Since the experimental uncertainty in microwave field strength is $\pm 10\%$, these deviations are not very significant. In the intermediate frequency region, both quantum numerical and experimental values are usually larger than classical ones, signs of a tendency for localization. The broad classical structures near $\omega_0 = 1$ and 2 are resonances, with only the former apparently playing a large role under present experimental conditions. For $\omega_0 > 0.8$, the classical results are well above the classical chaos border primarily because the microwave pulse time was less than the time for complete classical ionization $T_I \sim \omega_0^{7/3}/2F_0^2$.[7]

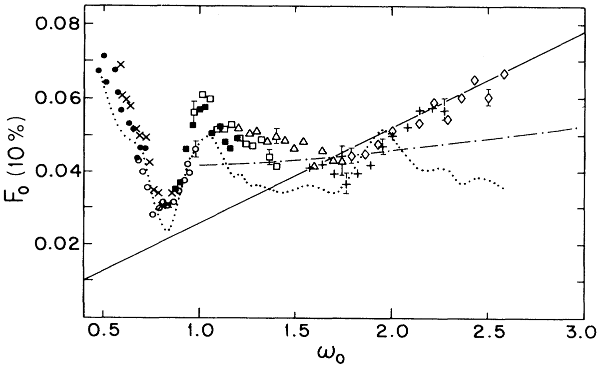

FIG. 2. Experimental data as in Fig. 1, compared with the results of one-dimensional model classical calculations (dotted line) and with one-dimensional model predictions for quantum suppression by cantori in phase space (dash-dotted curve). The solid line of Fig. 1 is also included. The dotted line was created by drawing through every point obtained for the parameter values used for the solid triangles of Fig. 1.

Also included in Fig. 2 is a smooth curve for the ionization threshold obtained from a model for quantum suppression arising from cantori in phase space just above the threshold for chaos.[18] This curve wave obtained from an analytical expression that was evaluated for the present experimental values of the parameters, with the choice of the typical microwave frequency 15 GHz. Although this curve very roughly follows the trend of the data between $\omega_0 = 1$ and 2, at higher frequencies it clearly departs from our experimental curve. Above $\omega_0 = 2$ the experimental data rise with ω_0 2.5 times faster than that predicted by the cantori model. In addition, numerical quantum calculations up to $\omega_0 = 4$ also show the more rapid rise.[13]

The present quantum calculations were based upon the one-dimensional model and employed numerical techniques previously described.[7,13] The procedure for dealing with the $n = 165$ static-field ionization threshold for the field in the waveguide region was as follows. As the microwave pulse rose towards its peak value, there was a time delay for a probability greater than 0.1% to arrive at the level $n = 165$, which typically was about 40 field oscillations. After a further time ΔT about equal to the Kepler period at $n = 165$, all population above $n = 164$ was removed from the system of coupled-state equations. Before doing this, it was checked that the removed population had not reached up to the $n = 365$ top of the hydrogenic basis set. This population removal procedure was contained for the rest of the microwave pulse. Calculations were carried out for various values of ΔT, with the finding that final threshold fields were independent of ΔT over a range of a factor of 2.

In conclusion, experimental conditions have been found where for classically scaled microwave frequencies $\omega_0 \gtrsim 2$, both experimental data and the numerical predictions of quantum mechanics agree with the analytical theory of localization and disagree with classical theory. The classical computations utilized the one-dimensional approximation, but the failure of three-dimensional classical theory in the present range of ω_0 has just been independently established experimentally.[19] Unlike the long-pulse experiments at twice our unscaled frequency ω,[19] the present results distinguish between the predictions of the localization and cantori models. A comparison of the two experimental results suggests that the cantori model's dependence of ω is not the correct one.

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