Classical Scaling of Nonclassical Stability in Microwave Ionization of Excited 3d H Atoms

B. E. Sauer, M. R. W. Bellermann, and P. M. Koch

Physics Department, State University of New York at Stony Brook, Stony Brook, New York 11794-3800

(Received 16 December 1991)

At each of three frequencies $\omega/2\pi=26.43$, 30.36, and 36.02 GHz, we observe 3d hydrogen atoms with certain principal quantum numbers $n_0 \gg 1$ to be more stable against microwave ionization. Which n_0's are more stable differs for each ω. Even when the enhanced stability is not found in 3d classical calculations, we show that it scales classically: This quantal effect occurs at certain values of the classically scaled frequency $n_0^3\omega$. Our data give strong support for the enhanced stability being due to scarred 3d wave functions for the strongly driven atoms.

PACS numbers: 05.45.+b, 03.65.−w, 32.80.Rm

Under active scrutiny are quantal systems whose low-dimensional, deterministic classical counterparts are chaotic. Many studies [1-3] have involved model systems that are either time independent $\overline{T}$ (spectral problem) or time dependent T (especially periodic, which are quasienergy spectral problems) and either closed C (bounded) or open O (dissociating or scattering). Here we focus on real H atoms [4-6] driven by a linearly polarized electric field whose amplitude F is comparable to the Coulomb binding field $F_{\rm at}$ for initial principal quantum number $n_0 \gg 1$. This is the paradigm for a periodically driven quantal system whose classical TO counterpart is chaotic [7].

In this Letter we demonstrate classical scaling of nonclassical effects in microwave "ionization" and argue, for the first time for *driven 3d H atoms*, that scarred wave functions [3,8,9] play an important stabilizing role [9]. [d (D) refers to configuration- (phase-) space dimension.] Our raw data are ionization curves, each recording the ionization probability $P_{\rm ion}$ vs F for a given n_0, ω, pulse-shape $A(t)$, and cutoff n value, n_c. Here "ionization" means true ionization plus excitation to bound states $n > n_c$ [4].

Though periodic orbits (PO) are of negligible measure in chaotic systems, they play a crucial role in semiclassical theories of their quantal counterparts [1-3,8]. For example, in a phase space divided by Kolmogorov-Arnol'd-Moser (KAM) tori (or cantori, broken KAM tori) into regular and chaotic regions [3,10], nonlinear trapping resonances form around stable PO. Since 1985 [11], data from our laboratory have demonstrated that these resonances can stabilize real 3d H atoms; we call this effect classical local stability (CLS).

That wave functions may be associated with *unstable* PO and other phase-space structures [12] was found in numerical studies of classically chaotic models. Extending earlier work [13], Heller [8] computed configuration-space wave functions for many excited states of the $\overline{T}C$ stadium billiards and graphically found their densities usually to peak along one or more (highly) unstable PO. He coined the term "scar" (wave function scarred by PO) to describe this surprising semiclassical phenomenon [3].

Earlier 36.02-GHz ionization data [4] from our laboratory showed that 3d H atoms can have nonclassical local stability (non-CLS). For example, $n_0=62$ was harder to ionize than its neighbors $n_0=61,63$, but 3d classical calculations showed no such effect. Within a 1d model of the atom, Jensen *et al.* [9] found the non-CLS to be caused by a scarred wave function prepared by the slow turn-on of the 36.02-GHz amplitude.

We designed the present experiments to test with 3d H atoms the scar-induced stability mechanism. Unable to measure wave functions [14], we investigated whether the non-CLS would scale classically. Therefore, we now discuss classical scaling [15].

Ignoring negligible spin and relativistic effects, the Hamiltonian in atomic units a.u. ($e=\hbar=\mu=1$, where μ is the reduced electron mass) for the driven H atom is $\mathcal{H}(t)=\mathbf{p}^2/2-|\mathbf{r}|^{-1}+zA(t)F\sin(\omega t+\varphi)$, where $0 \le A(t) \le 1$ is slowly varying, and φ is an initial phase averaged by the experiment. [Because linear polarization preserves the projection of angular momentum (quantally, $|m|$) on the field axis, the motion is confined to a 5D surface in the extended [10] phase space. Quantally, one needs solutions of the 2d time-dependent Schrödinger equation for each value of $|m|$.] For an arbitrary constant we choose to be n_0, under the similarity transformation [$\tilde{\mathbf{p}}=n_0\mathbf{p}$, $\tilde{\mathbf{r}}=\mathbf{r}/n_0^2$, $\tilde{t}=t/n_0^3$, $\tilde{\varphi}=\varphi$], $\mathcal{H}(t)$ becomes

$$\tilde{\mathcal{H}}(\tilde{t})=n_0^2\mathcal{H}(t)=\tilde{\mathbf{p}}^2/2-1/|\tilde{\mathbf{r}}|+\tilde{z}A(n_0^3\tilde{t})\tilde{F}\sin(\tilde{\omega}\tilde{t}+\tilde{\varphi}) . \tag{1}$$

$\tilde{\omega}=n_0^3\omega\equiv\Omega_0$ is the scaled frequency [16], the ratio of ω and the initial Kepler orbital frequency; $\tilde{F}=n_0^4F\equiv F_0$ is the scaled amplitude, the ratio of F and $F_{\rm at}$.

When $A(t)=$const, the classical dynamics [9,15,17] depends on Ω_0 and F_0, not separately on n_0, ω, and F. The quantum dynamics is different because the similarity transformation does not preserve the canonical commutation relations. For example, we interpret $[\tilde{x},\tilde{p}_x]=i\hbar/n_0$ as giving an effective $\hbar$, $\tilde{\hbar}=\hbar/n_0$.

For a given $A(t)\neq$const, exact classical scaling would require varying the interaction time as n_0^3, i.e., for a fixed number of initial Kepler periods. Experimentally, one may demonstrate classical scaling if different values of n_0, ω, and F produce the same ionization behavior when

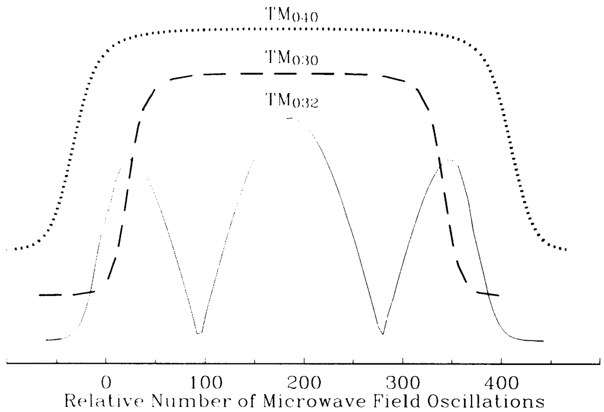

FIG. 1. Absolute value $|A(t)|$ of the experimental pulse shape, displaced vertically for clarity, for each of three microwave frequencies: Top, 36.02 GHz; middle, 26.43 GHz; bottom, 30.36 GHz.

Ω_0 and F_0 are the same, even if $A(t)$ is not scaled exactly. Figure 7 of [5] demonstrates this with $n_0=74$ 3d atoms in a 9.92-GHz field ($\Omega_0=0.611$) and $n_0=48$ 3d atoms in a 36.02-GHz field ($\Omega_0=0.605$); $\tilde{\hbar}$ was varied by 54%.

The present apparatus differs only slightly from that described previously [4,5]; here we emphasize important details. A 14.6-keV beam of H atoms, initially in a uniform distribution of substrates belonging to a fixed n_0 value prepared by double-resonance laser excitation, traversed a cylindrical microwave cavity [18] whose symmetry axis coincided with the beam axis. Holes through the end caps for beam entry and exit affected the cavity eigenmodes. For each of three modes we determined to 5% accuracy [19] the amplitude F [18] seen by the atoms. We numerically calculated [20], and experimentally confirmed with a "bead-pull" resonance-frequency-perturbation method [21], the spatial distribution of F. In the atomic rest frame, each spatial distribution became an $A(t)$. Figure 1 shows similar *initial rising* slopes and *final falling* slopes for all three $A(t)$ functions, but we expect microwave ionization at 30.36 GHz to be dominated by the largest (center) antinode, whose shape (half of a sinusoid) and duration [full width {width at amplitude $>95\%$} $=182$ {39} field oscillations] are quantitatively similar to those in other H atom experiments with a 12–18-GHz waveguide interaction region [6].

As was described in [4], electron ionization and proton quench signals, which differed mainly in their n cutoffs, n_c^i and n_c^q, respectively, were detected simultaneously with different particle multipliers and stored in a computer. $n_c^i \simeq 114$ was determined by an axial static-electric field $F_{dc}=3.4$ V/cm (entirely outside the microwave field) between the exit endcap and a planar electrode placed 1.1 cm after the cavity. Other static fields downstream determined the value of $n_c^q \simeq 95$. The static electric field *inside* the cavity was $\lesssim 0.017$ V/cm; it could ionize atoms with

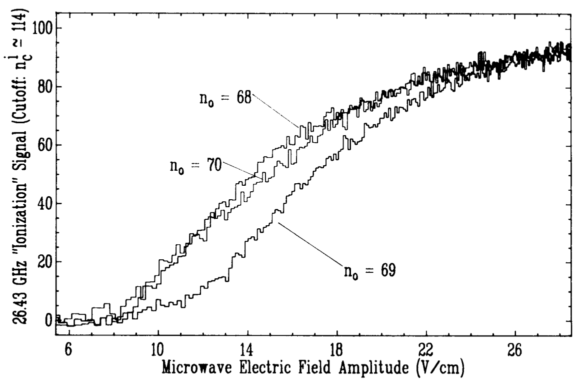

FIG. 2. Measured "ionization" curves for $n_0=68,69,70$ 3d H atoms in a 26.43-GHz field, with the middle pulse shape shown in Fig. 1.

$n \gtrsim 440$, far above n_c^i and n_c^q.

In Fig. 2, $n_0=69$ ($\Omega_0=1.32$) is significantly more stable against ionization at 26.43 GHz than either neighboring n_0 value. At 30.36 and 36.02 GHz, respectively, we observed similar behavior for $n_0=66$ ($\Omega_0=1.33$) and for $n_0=62$ ($\Omega_0=1.31$).

From each of many ionization curves we obtained $X\%$ thresholds, the amplitudes $F(X)$ that produce $P_{ion}=X\%$. Figure 3 plots scaled 10% thresholds $F_0(10)$ vs Ω_0 for all three frequencies as well as those obtained from a classical 3d Monte Carlo simulation [4] of the 36.02-GHz experiment, including its $A(t)$ and $n_c^i \simeq 114$. All results show a local maximum near $\Omega_0=1$, a nice example of CLS reflecting the main classical nonlinear trapping resonance. As will be discussed below, the less rapid falloff of the 30.36-GHz data just above $\Omega_0=1$ is probably a result of its very different $A(t)$ function; see Fig. 1. Its much shorter near-peak-amplitude interaction time likely explains why the 30.36-GHz $F_0(10)$ thresholds are systematically the highest measured.

The local maxima near $\Omega_0=1.3$ are the most prominent example of non-CLS in Fig. 3. *Even though it is*

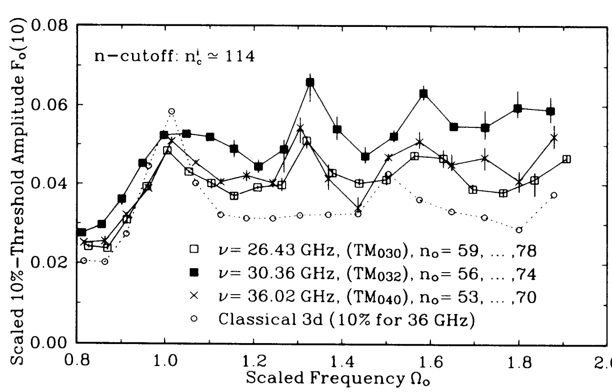

FIG. 3. Scaled 10% thresholds vs scaled frequency. All cases had the n cutoff $n_c^i \simeq 114$ determined by a 3.4-V/cm static field. Straight lines join points in each data set.

absent in classical calculations, the non-CLS scales classically: At, just above, and just below $\Omega_0 \simeq 1.32$, the $F_0(10)$ values are quite close for 26.43 and 36.02 GHz, which had similarly shaped $A(t)$ functions. More surprising is the large local maximum for 30.36 GHz; even with its $A(t)$ function, the non-CLS effect is preserved. Similar remarks apply to another example of non-CLS apparent in Fig. 3, just below $\Omega_0 = 1.6$. This is *above* $\Omega_0 = \frac{3}{2}$, where there is a classical nonlinear resonance.

Note that overlaying unscaled plots (not shown here) of $F(10)$ in V/cm vs n_0 for the three frequencies shows none of the detailed coincidences present in the scaled plot in Fig. 3.

We now discuss our non-CLS data in the light of scarred wave functions. Jensen *et al.* [9] linked the stability of $n_0 = 62$ in a 1d model H atom in a strong 36.02-GHz field ($F_0 = 0.05$) to its wave function being dominated (98%) by one quasienergy eigenfunction prepared by the slow rise in $A(t)$. When projected onto Poincaré sections in the 2D classical phase space, the Husimi distribution of this dominant eigenfunction was located *outside* the main nonlinear resonance centered near $\Omega_0 = 1$ and was concentrated in the chaotic region near an unstable PO (hyperbolic fixed point) and associated stable and unstable manifolds [9]. It was a scar [3,8]. Neighboring initial states, $n_0 = 61,63$, were less stable because their strong-field 1d wave functions contained appreciable amplitudes of quasienergy eigenfunctions that connected to much higher lying, more easily ionized atomic states. These results [9] were for H atoms in a 1d model.

It is not at all obvious *a priori* that there would be scar-induced stability for real 3d H atoms. Though previous 2d and 3d classical calculations say nothing about wave functions, they show [17] the following: (i) $F_0(10)$ values for 3d H atoms are usually well approximated by the 1d model for $\Omega_0 \lesssim 3$. Very-low-$|m|$ orbits are stretched into nearly 1d orbits and ionize at the lowest amplitudes. (ii) 1d calculations seriously underestimate values of F_0 needed to reach higher values of $P_{\rm ion}$, to which much higher-$|m|$ 3d orbits contribute. In addition, some numerical calculations and an iterated (Kepler) map showed [22] that a quantal dynamical localization effect present in the 1d dynamics was preserved in 2d, at least for Ω_0 between 1 and 2.5.

Let us assume that given n_0, ω, and $A(t)$, X% ionization thresholds rise monotonically with increasing $|m|$, which is bounded by $n_0 - 1$. A calculation then shows that each value of $P_{\rm ion}$ is equal to the fraction f of atoms with $|m| \leq |m_c(f)| = \{2n_0 - 1 - [4n_0^2(1-f)+1]^{1/2}\}/2$. For $n_0 = 69$, for $P_{\rm ion} = 10\%$ ($f = 0.1$), $|m_c| \simeq 3$, a small value; for $P_{\rm ion} = 80\%$ ($f = 0.8$), $|m_c| \simeq 38$, a large value. The enhanced stability of $n_0 = 69$ at 26.43 GHz for values of $P_{\rm ion}$ up to at least 80% in Fig. 2 was also observed for $n_0 = 62$ at 36.02 GHz and for $n_0 = 66$ at 30.36 GHz. Thus, the non-CLS for $\Omega_0 \simeq 1.32$ scales classically, even for high $P_{\rm ion}$ values. Our data also show similar behavior for Ω_0 just below 1.6. *We interpret these results as strong evidence for the stabilizing effect of scarred wave functions in the driven 3d H atom*: These wave functions must "live" on structures in the 5D phase space. They need to be found, visualized, and understood.

Looking in greater detail at Fig. 2, note that the $n_0 = 69$ ionization curve starts to rise with the $n_0 = 68,70$ curves but then levels off at $P_{\rm ion} \simeq 5\%$ between about 9 and 11 V/cm. We infer that the strong-field wave function for $n_0 = 69$ contained about a 5% admixture of component(s) that ionized more easily than the dominant, scarred component. Recall that two different quantal 1d calculations [9,23] found such a (2–3)% admixture for an $n_0 = 62$ 1d model atom in a 36.02-GHz field whose $A(t)$ function was similar to the flattop one(s) in Fig. 1.

Remarkably, that we observed similar $\simeq 5\%$ plateaus (not shown) for $n_0 = 62$ at 36.02 GHz and $n_0 = 66$ at 30.36 GHz suggests classical scaling for this phenomenon. Moreover, that the entire ionization curves for the three cases were unaffected by increasing the cutoff from $n_c^q \simeq 95$ to $n_c^i \simeq 114$ shows that the excitation was to final states $n \gtrsim 114$. (It was probably true ionization [4].) For other (non-scar-dominated) initial states, changing n_c could produce a significant change in their "ionization" curves.

For 1d quasienergy states, Breuer and Holthaus [24] found an approximate scaling relation that is broken by quantal avoided crossings. As $A(t)$ varies the amplitude, how these avoided crossings are traversed (i.e., the Landau-Zener mechanism) undoubtedly affects $P_{\rm ion}$. Because they neglected the continuum, however, the calculations [23,24] said nothing directly about true ionization, only about excitation and deexcitation. The calculations also investigated [23] how quickly the $A(t)$ function would have to turn on for a 1d wave function no longer to be dominated by a stable, "continuously connected" quasienergy state. For 1d $n_0 = 62$ at 36.02 GHz, the time scale was about five field oscillations; for 1d $n_0 = 57$ at 36.02 GHz, very near $\Omega_0 = 1$, where the main classical nonlinear resonance is centered, the time scale was about 10 times longer.

Extrapolating these 1d calculations [23,24] to 3d atoms suggests why our 30.36-GHz data differ in details from the 26.43- and 36.02-GHz data: The three $A(t)$ functions in Fig. 1 have comparable initial turn-ons and final turn-offs, but for 30.36 GHz, $A(t)$ has steeper slopes near $F = 0$ on either side of the center antinode.

Addressing more details, the 36.02-GHz ionization curves (not shown) for the trio $n_0 = 56,58,62$ are nearly *identical* over their whole range. The same is true for $n_0 = 61,65,69$ at 26.43 GHz and for $n_0 = 59,62,66$ at 30.36 GHz. Because the lower two members of each trio straddle the $\Omega_0 = 1$ resonance, their strong-field wave functions are presumably associated with an Einstein-Brillouin-Keller-type quantization on invariant tori [3,9]. The highest member is the scarred state at $\Omega_0 \simeq 1.32$. That all members of each trio have the same ionization curve reinforces the picture that the 3d quantization of the scarred state is, indeed, associated with the classical $\Omega_0=1$ resonance. Conversely, we do not yet know the corresponding association for the non-CLS just below $\Omega_0=1.6$.

Experiments at Pittsburgh with $n_0=80,86$ and $\omega/2\pi$ between 12 and 18 GHz did not find non-CLS near $\Omega_0=1.3$; see Fig. 2 in [6]. The cutoff there, $n_c\simeq 1.5n_0$, was similar to the present n_c^i. Given the non-CLS in our 30.36-GHz data with 3d atoms, we think it unlikely that their nonobservation of non-CLS is due to their $A(t)$ function or quasi-2d substrate distribution [6]. There was, however, an $F_s=0.87$ V/cm static field inside their microwave interaction region [6]. No calculations evaluating its effect on the scar stability have been reported, but the effect of an F_s on the 1d spectrum of quasienergy states has been discussed [24]. Our preliminary results (not shown) suggest that a static field of 1 V/cm affects the non-CLS of $n_0=69$ in the 26.43-GHz field. The effect of extra perturbations (static fields, other frequencies, and, especially, noise) on the scarred states clearly needs to be investigated.

Finally, for non-CLS at Ω_0 near 1.3 (and 1.6), we have varied $\tilde{h}$ by only 11%. What will happen for much wider variations? For, say, an $n_0=2$ H atom in an optical laser field, we would not expect non-CLS: The effective $\hbar$ should be too large for scarring. For, say, an $n_0\simeq 124$ atom in a 4.50-GHz field (and a much higher n_c), $\tilde{h}$ would be 2 times smaller than at present; one might expect scarring to affect two neighboring values of n_0. However, this cannot continue to the $n_0\rightarrow\infty$ limit: The 3d classical calculations in Fig. 3 show no enhanced stability near $\Omega_0=1.3$. The enhanced stability here is a *semiclassical* effect that must disappear in the deep quantal limit *and* in the classical limit [25]. It is a splendid example of the subtlety of quantum chaology [26].

We thank M. V. Berry, R. V. Jensen, D. Richards, and B. Sundaram for discussions and L. Moorman for much help in beginning these experiments. We appreciate financial support from the NSF.

[1] See papers in *Chaos and Quantum Physics*, edited by M.-J. Giannoni, A. Voros, and J. Zinn-Justin (Elsevier, Amsterdam, 1990).

[2] See papers devoted to *Irregular Atomic Systems and Quantum Chaos*, edited by J.-C. Gay [Comments At. Mol. Phys. **25**, Nos. 1-3, 4-6 (1990)].

[3] A. M. Ozorio de Almeida, *Hamiltonian Systems: Chaos and Quantization* (Cambridge Univ. Press, Cambridge, 1988); M. C. Gutzwiller, *Chaos in Classical and Quantum Mechanics* (Springer, New York, 1990).

[4] E. J. Galvez, B. E. Sauer, L. Moorman, P. M. Koch, and D. Richards, Phys. Rev. Lett. **61**, 2011 (1988).

[5] P. M. Koch, L. Moorman, B. E. Sauer, E. J. Galvez, K. A. H. van Leeuwen, and D. Richards, Phys. Scr. **T26**, 51 (1989).

[6] J. E. Bayfield, G. Casati, I. Guarneri, and D. W. Sokol, Phys. Rev. Lett. **63**, 364 (1989).

[7] T. Tél, in *Directions in Chaos*, edited by Hao Bai-lin (World Scientific, Singapore, 1990), Vol. 3, discusses "transient chaos."

[8] E. J. Heller, Phys. Rev. Lett. **53**, 1515 (1984); E. J. Heller, P. W. O'Connor, and J. Gehlen, Phys. Scr. **40**, 354 (1989).

[9] R. V. Jensen, M. M. Sanders, M. Saraceno, and B. Sundaram, Phys. Rev. Lett. **63**, 2771 (1989); R. V. Jensen, S. M. Susskind, and M. M. Sanders, Phys. Rep. **201**, 1 (1991).

[10] A. J. Lichtenberg and M. A. Lieberman, *Regular and Stochastic Motion* (Springer, New York, 1983); B. V. Chirikov, Phys. Rep. **52**, 263 (1979).

[11] 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).

[12] G. Radons and R. E. Prange, Phys. Rev. Lett. **61**, 1691 (1988).

[13] S. W. McDonald and A. N. Kaufman, Phys. Rev. Lett. **42**, 1189 (1979).

[14] S. Sridar, Phys. Rev. Lett. **67**, 785 (1991).

[15] J. G. Leopold and I. C. Percival, J. Phys. B **12**, 709 (1979).

[16] P. M. Koch, L. Moorman, and B. E. Sauer in [2] delineated with different ranges of $n_0^3\omega$ six regimes of dynamical behavior in microwave ionization.

[17] D. Richards, in *Aspects of Electron-Molecule Scattering and Photoionization*, edited by A. Herzenberg, AIP Conf. Proc. No. 204 (AIP, New York, 1990); D. Richards, in [2].

[18] Cavity: 2.007-cm-length, 3.130-cm-diam, 0.64-cm-thick end caps having 0.258-cm-diam axial holes. For our TM modes, F_z decreased quadratically with ρ by a few percent from $\rho=0$ to the $\rho_e=0.07$-cm radial edge of the collimated atomic beam. All values of F in this Letter refer to a "median" value evaluated at $\rho_e/\sqrt{2}=0.049$ cm.

[19] B. E. Sauer, K. A. H. van Leeuwen, A. Mortazawi-M, and P. M. Koch, Rev. Sci. Instrum. **62**, 189 (1991).

[20] K. Halbach and R. F. Holsinger, Part. Accel. **7**, 213 (1976).

[21] L. C. Maier and J. C. Slater, J. Appl. Phys. **23**, 68 (1954).

[22] G. Casati, I. Guarneri, and D. L. Shepelyansky, IEEE J. Quantum Electron. **24**, 1240 (1988).

[23] H. P. Breuer, K. Dietz, and M. Holthaus, Z. Phys. D **18**, 239 (1991).

[24] H. P. Breuer and M. Holthaus, Ann. Phys. (N.Y.) **211**, 249 (1991).

[25] See also, D. Wintgen and A. Hönig, Phys. Rev. Lett. **63**, 1467 (1989), who theoretically found the collective importance of wave-function scarring for the H atom in a strong magnetic field to disappear as $h\rightarrow 0$.

[26] M. Berry, Phys. Scr. **40**, 335 (1989).