Effect of Closed Classical Orbits on Quantum Spectra: Ionization of Atoms in a Magnetic Field

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Measurements of the absorption spectrum near the ionization threshold for an atom in a strong magnetic field showed that the spectrum is a superposition of many oscillatory terms ("quasi-Landau oscillations"). We have developed a quantitative theory which shows that each classical closed electron orbit which begins and ends near the nucleus contributes an oscillatory term to the average oscillator strength. The theory gives new understanding of the behavior under combined Coulomb and Lorentz forces, and it elucidates the roles of isolated closed orbits in chaotic systems. The first results of this theory are shown to be in good agreement with experimental results.

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The relationship between classical mechanics and quantum mechanics is rather well understood for integrable systems. Such systems admit a set of conserved classical action variables, and the energy eigenvalues correspond closely to trajectories having appropriately quantized values of these action variables. For systems in which the classical motion is irregular or chaotic, on the other hand, the quantum behavior is poorly understood, and the whole field of "quantum chaos" is marked by confusion and controversy.[1] It is easier to identify properties which chaotic systems lack (e.g., conservation laws) than the properties they possess.

There is evidence that in such systems, when the density of states gets very high, eigenfunctions and eigenvalues become very unstable under small changes in the calculational method.[2] This would mean that these most fundamental quantities may be exceedingly difficult to calculate and to measure. If this is correct, then the major problem is to identify properties which can be calculated and measured, and which in this sense constitute stable attributes of the system.

Important insight comes from experimental measurements of the absorption spectrum of atoms near the ionization threshold. If the atom is in field-free space, then the observed oscillator strength is a smooth and slowly varying function of photon energy, going continuously from a finite value above threshold to the same average value below threshold. Almost twenty years ago, Lu, Tomkins, and Garton[3] showed that if the atom is placed in a magnetic field, then the absorption spectrum shows sinusoidal oscillations superimposed on this smooth background. Edmonds[4] pointed out that these oscillations are correlated with a periodic orbit in the system.[5]

Recently the near-threshold spectrum of hydrogen atoms in magnetic fields has been measured with much improved resolution.[6] It was found that the observed oscillator strength is in fact a superposition of many sinusoidal oscillations. Furthermore, the "wavelength" (or peak-to-peak energy spacing ΔE_n) of each oscillation corresponds to the period T_n of a classical periodic orbit of the system through the relationship $\Delta E_n = 2\pi\hbar/T_n$.

Computational evidence indicates that these systems are classically chaotic, with only isolated, unstable periodic orbits. Why do these orbits produce such phenomena?[7]

Many years ago Gutzwiller[8] proved that periodic classical orbits produce oscillations in the density of states of a quantum system. However, spectral measurements do not directly observe the state density but rather the average oscillator-strength density, $\overline{Df}(E)$: the transition dipole moment averaged over the small range of energy corresponding to the experimental resolution,[9]

$$\overline{Df}(E) = \left(\frac{2m}{\hbar^2}\right) \int |\langle \psi_f | D | \psi_i \rangle|^2 (E_f - E_i)\rho(E_f) g(E_f - E) dE_f. \quad (1)$$

We report here the development of a quantitative theory which shows the relationship between closed orbits and the observed oscillations in the spectrum.

The theory and calculations are based upon two approximations. (1) Close to the nucleus ($r \lesssim 50a_0$), the effect of the magnetic field is neglected, and the electron wave function corresponds to zero-energy scattering in a Coulomb field. (2) Far from the nucleus ($r \gtrsim 50a_0$) a semiclassical approximation is used. These approximations lead to a simple physical picture.

When the atom absorbs a photon, the electron goes into a near-zero-energy Coulomb outgoing wave. This wave propagates away from the nucleus to large distances. For $r \gtrsim 50a_0$ the outgoing wave fronts propagate according to semiclassical mechanics, and they are correlated with outgoing classical trajectories. Eventually the trajectories and wave fronts are turned back by the magnetic field; some of the orbits return to the nucleus, and the associated waves (now incoming) interfere with the outgoing waves to produce the observed oscillations.

From these ideas, and with these approximations, we can show that the observed oscillator strength can be written as a smooth, slowly varying background term plus a sum of sinusoidal oscillations:

$$\overline{Df}(E)=Df_0(E)+\sum_n A_n(E)\sin\left[\int_0^E T_n(E')dE'+\alpha_n\right]. \quad (2)$$

The background term $Df_0(E)$ is precisely the oscillator strength density that would be obtained in the absence of an external field.

Each oscillatory term corresponds to a closed orbit of the electron in the combined Coulomb and magnetic fields. Each closed orbit begins and ends at the atomic nucleus.[10] $T_n(E)$ is the transit time for the electron on this orbit; it is a slowly varying function of E (in most cases essentially constant over the relevant range of E). If the spectrum is measured at low resolution, then only the orbits of shortest duration contribute to this sum; orbits of longer duration produce rapidly oscillating terms that average to zero. With increasing resolution, more and more terms become significant, and the spectrum is found to oscillate wildly.

The amplitudes of the oscillations, $A_n(E)$, depend upon (1) the initial state of the system; (2) the polarization of the absorbed light; (3) the initial and final directions of the orbit as it leaves and returns to the nucleus; and (4) the relative stability of the closed orbit, i.e., the divergence of adjacent trajectories from the central closed orbit.

The phase constant α_n for each oscillatory term is also related to the initial state, light polarization, and initial and final directions; in addition, it is related to the classical action integral $\int \mathbf{p}\cdot d\mathbf{q}$ on the orbit at zero energy, and it contains Maslov phase corrections associated with caustics or focal points through which the orbit passes.

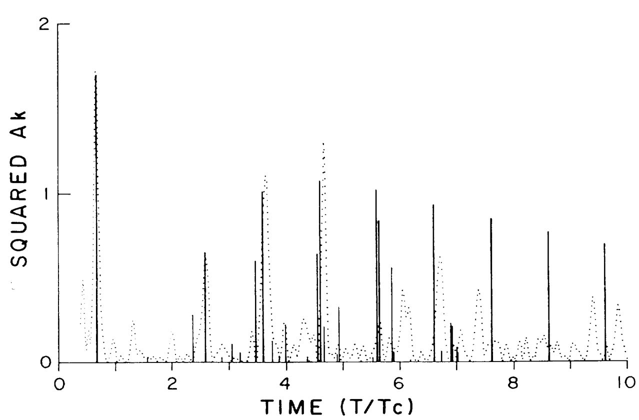

FIG. 1. Fourier-transformed spectrum. Vertical lines show calculated amplitudes $|A_n|^2$ vs orbit durations T_n. Dotted line is experimental result from Ref. 5. The experimental result was normalized to match the height of the largest peak (T/T_c =0.67).

A complete set of formulas for these quantities and the derivation of these formulas will be presented in a future publication. Here we show some of our first results that can be compared with experiment. Because the spectrum itself is wildly oscillatory, a direct comparison between theoretical and experimental oscillator strengths is unhelpful. More appropriate for comparison is the Fourier transform of the spectrum, which was obtained by Main *et al.*[11] We show their result compared to our calculated amplitudes in Fig. 1. Very pleasing agreement is obtained for the short-period orbits ($T/T_c \lesssim 4.6$).[12]

The phases α_n are more difficult to calculate to acceptable accuracy than are the periods or amplitudes. The classical action integrals are large numbers (the smallest is $\sim 250\hbar$) so that if the semiclassical approximation were to produce a small percentage error in α_n, then the calculated phase of the oscillations would be meaningless. We present a few of our calculated phases in Table I. Experimental values for these phases are not yet available.

One *apparent* point of disagreement between theory and experiment is particularly interesting. The theory shows the presence of two orbits (one periodic and one closed) having periods close to $5.6T_c$ which contribute oscillations with large amplitudes. The experiments show no significant peak there. In fact the phases of these two oscillatory terms differ by nearly π, so that the two terms almost exactly cancel. Hence theory and ex-

TABLE I. Trajectory and spectrum data. Values listed here are the largest ten amplitude oscillations calculated for transition from $2p_z$ to $m_f=0$ and $E_f=0$ at $B=5.96$ T.

A[a] (hartree^{-1})	T[b]	α[c]	$-d\alpha/dB$ (T^{-1})	θ_i[d]	N[e]
1.31	0.666	5.30	13.9	90.0000[f]	1
1.04	4.596	0.98	29.8	33.8359	9
1.01	3.589	5.47	27.4	37.3112	7
1.01	5.602	3.01	31.8	31.3650	11
0.97	6.605	0.77	33.6	29.4816	13
0.92	5.627	6.05	42.9	76.0617[g]	13
0.92	7.608	1.65	35.3	27.9784	15
0.88	8.611	0.02	36.8	26.7390	17
0.83	9.612	2.65	38.1	25.6917	19
0.81	2.579	0.49	24.5	42.8096	5

[a]Usually contributions come from two trajectories, one being the mirror image of the other about the $z=0$ plane.
[b]In period of cyclotron motion, $T_c=6.0\times10^{-12}$ s.
[c]Mod(2π).
[d]Initial polar angle of trajectories from the z axis.
[e]Number of passages through a caustic or focus in (ρ,z) space.
[f]This trajectory counts only once.
[g]This is a closed, not periodic orbit; the final angle is 146.9510°. It counts 4 times (reflection about $z=0$, and time reversal).

periment actually agree at this point. This suggests that our phase calculations might be quite accurate, and it establishes that not only periodic but also closed orbits must be included in the sum (2).

To conclude, we have shown that stable and orderly properties of a quantum chaotic system are associated with closed classical orbits of the system. In hindsight this is not surprising. Chaos in classical Hamiltonian systems is a characteristic of the behavior of the system in the limit $T \to \infty$. Observations made over short periods of time do not necessarily show evidence of chaotic behavior. In quantum mechanics, eigenfunctions and eigenvalues are stationary properties of a system, and they are correlated with the behavior of orbits over infinite (or at least very long) times. However, a low-resolution spectrum involves shorter-time phenomena. In the present case, the spectrum shows orderly patterns correlated with the orderly behavior of the classical trajectories for restricted periods of time.

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[1]See, for example, *Chaotic Behavior in Quantum Systems,* edited by G. Casati (Plenum, New York, 1985), and Proceedings of the First International Conference on the Physics of Phase Space, College Park, Maryland, 1986, edited by W. W. Zachary (to be published).

[2]I. C. Percival, Adv. Chem. Phys. **36**, 1 (1977); C. W. Clark and K. T. Taylor, J. Phys. B **15**, 1175 (1982); M. V. Berry, in *Chaotic Behavior of Deterministic Systems,* edited by G. Iooss, R. H. G. Helleman, and R. Stora (North-Holland, Amsterdam, 1983).

[3]K. T. Lu, F. S. Tomkins, and W. R. S. Garton, Proc. Roy. Soc. London, Ser. A **362**, 421 (1978).

[4]A. R. Edmonds, J. Phys. (Paris), Colloq. **31**, C4-71 (1970).

[5]An interpretation based upon Gaussian wave packets has been given by W. P. Reinhardt, J. Phys. B **16**, L635 (1983).

[6]A. Holle, G. Wiebusch, J. Main, B. Hager, H. Rottke, and K. H. Welge, Phys. Rev. Lett. **56**, 2594 (1986).

[7]Recall that in integrable systems, eigenvalues are correlated not with periodic orbits but with quasiperiodic orbits.

[8]M. Gutzwiller, J. Math. Phys. **12**, 343 (1971). See also M. V. Berry and M. Tabor, Proc. Roy. Soc. London, Ser. A **349**, 101 (1976).

[9]Here ψ_i and ψ_f are the initial and final states of the system with energies E_i and E_f, $\mathbf{D}$ is the dipole operator, $\rho(E_f)$ is the density of final states, and $g(E_f - E)$ is a smoothing or averaging function with width corresponding to the finite resolution of the experiment.

[10]The orbits are not necessarily periodic with period T; they may leave the nucleus in one direction and return from another.

[11]J. Main, G. Wiebusch, A. Holle, and K. H. Welge, Phys. Rev. Lett. **57**, 1789 (1986).

[12]At present we do not have any reason to believe that the theory is less accurate at large T/T_c. For the experimental values, large T corresponds to small ΔE, and $T/T_c \gtrsim 6$ might be approaching the limits of the experimental resolution.