Title
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SYNCHROTRON LIGHT SOURCES:
The search for quantum chaos

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ABSTRACT

A storage ring is a specialized synchrotron in which a stored beam of relativistic electrons circulates for many hours, producing broadband radiation which includes vacuum ultraviolet and x rays, depending on the energy of the electrons. A modern light source produces a bright photon beam, which can be focused to a small spot, and can thus fill the entrance slit of a monochromator; the result is a high flux of photons with high spectral brightness. One application for a photon beam of high spectral brightness has been to study doubly excited autoionizing states of the helium atom, which is a prototypical three-body atomic system. A group led by Günter Kaindl has performed experiment at the ALS to search for quantum chaos. New experimental results and theory indicate that the onset of quantum chaos has been observed.

Synchrotron light sources

The electromagnetic spectrum consists of radiation with photon energies (and wavelengths) over a very great range. Different sources are required to produce radiation in different energy ranges.

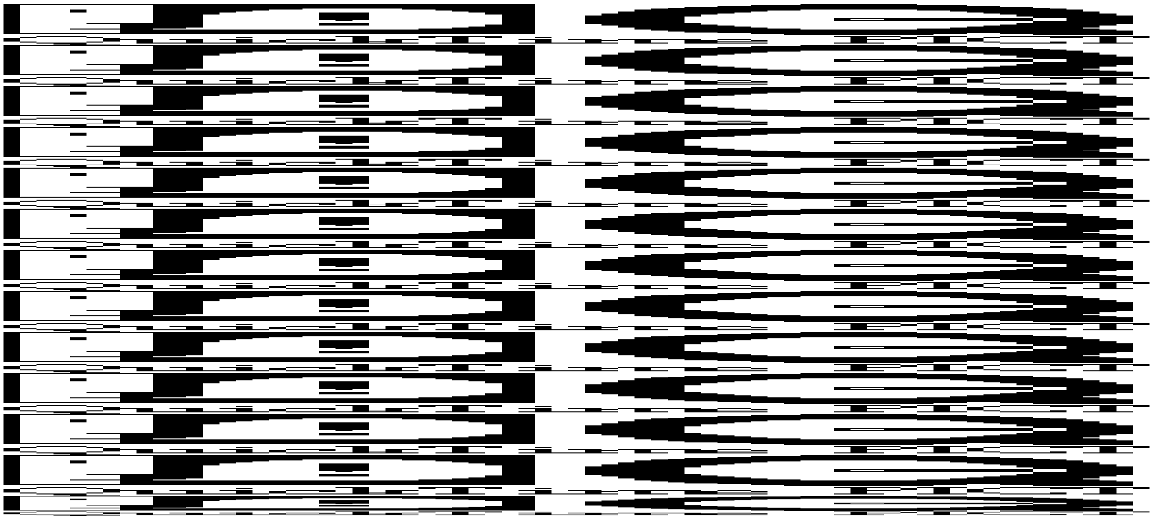


Figure 1. The electromagnetic spectrum. Radiation in the vacuum-ultraviolet and soft-x-ray regions of the spectrum is produced by a synchrotron light source like the ALS. (Figure courtesy of ALS.)

A storage ring enables a current of electrons to circulate at nearly the speed of light for many hours. Radiation is produced wherever the direction of motion of the electrons is changed, which occurs in dipole (bend) magnets and in undulators [1].

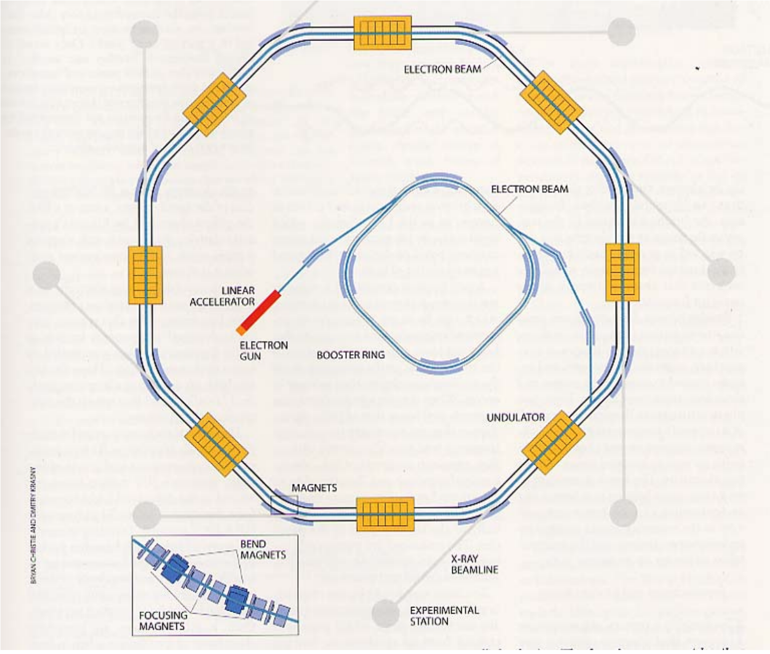


Figure 2. A storage ring enables a current of electrons to circulate at nearly the speed of light for many hours. Radiation is produced where the direction of motion of the electrons is changed [1].

X rays are of particular use in science because they interact selectively with electrons in matter; electrons determine most of the properties of interest of matter, such as magnetism, thermal and electrical conductivity, and many other properties.

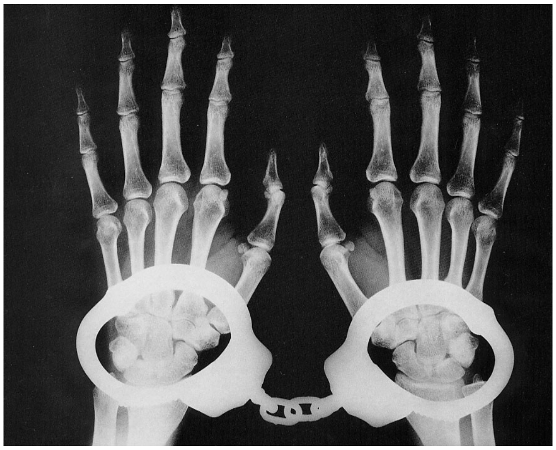


Figure 3. An x-ray image produced by differential absorption in matter. Higher density and higher atomic number cause more x rays to be absorbed, thus producing contrast in a shadowgram.

Radiation is produced in a storage ring by bend magnets and by specialized magnetic devices called undulators. An undulator causes a stored electron beam to bend back and forth many times over a length of a few meters. The light waves emitted at each bend overlap and either reinforce or cancel one another, depending on their wavelengths. The result is that certain wavelengths, or photon energies, are enhanced. Light at these wavelengths emerges in a narrow cone.

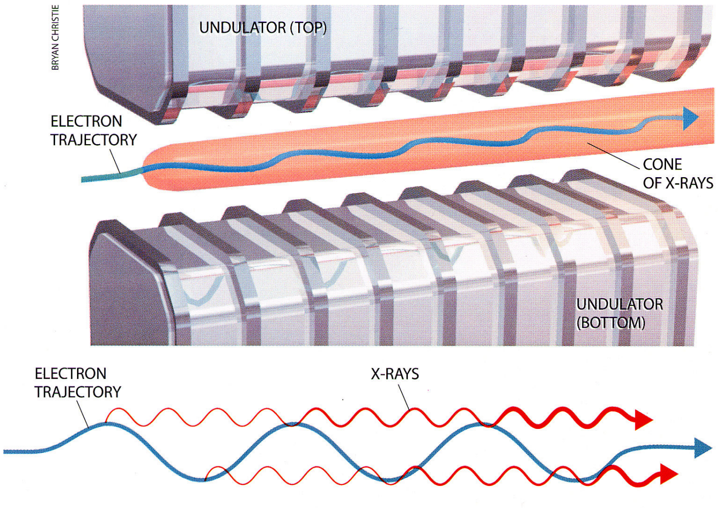


Figure 4. An undulator creates a spatially alternating magnetic field that bends electrons back and forth many times to produce an x-ray beam of exceptional brightness [1]. Waves from different points along the electron trajectory overlap one another because x rays are emitted in a narrow cone. Only waves of certain frequencies overlap one to produce constructive interference.

The spatial and spectral properties of radiation produced by bend magnets and by undulators are very different. Radiation from a bend magnet is white, i.e., broadband, and it has a large spatial size, filling the horizontal plane and with a height determined by relativistic kinematics. Radiation from an undulator consists of one or several harmonic peaks in energy, and is concentrated spatially in a narrow cone.

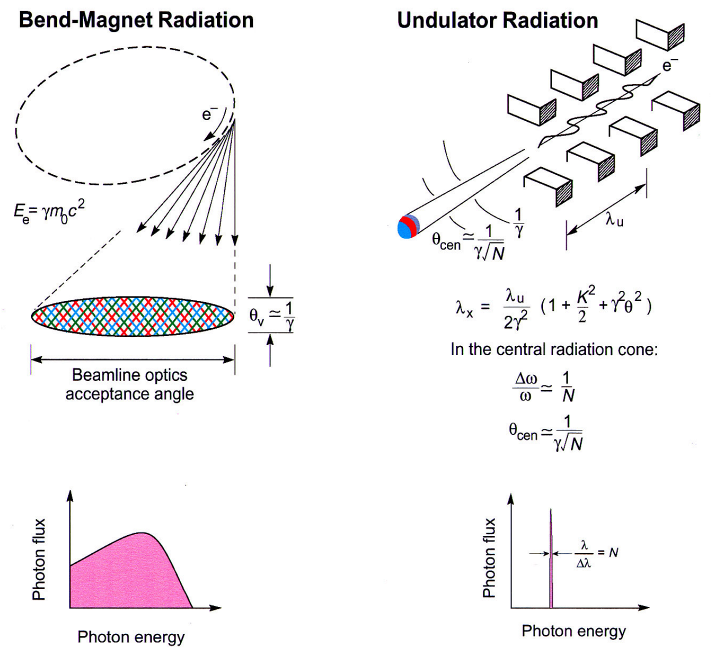


Figure 5. Spatial and spectral characteristics of radiation from a bend magnet and an undulator. (Figure courtesy of ALS.)

Synchrotron radiation has several properties of interest for research applications. For the Advanced Light Source in Berkeley, for example, the flux from an undulator can exceed 10^{13} photons/s with a spectral resolving power ($E/\Delta E$ = 10,000), or the beam can be focused to a very small spot. The radiation from an undulator is essentially 100% plane polarized, and circular and elliptically polarized radiation can be produced. The pulse repetition rate is 500 MHz (2 ns between pulses lasting around 30 ps) in normal multibunch operation; the time between pulses in special-operation two-bunch mode is approximately 300 ns.

The combination of a low-emittance lattice, undulators, and control of all parameters affecting the stability of the electron and photon beams has lead to a very great increase in the spectral brightness of modern storage rings compared to their predecessors. Brightness is density in phase space, or photon flux per unit source size, divergence, and bandwidth. High brightness allows a photon beam to be focused to a small spot, or to fill an entrance slit of a monochromator, and/or to be partially coherent.

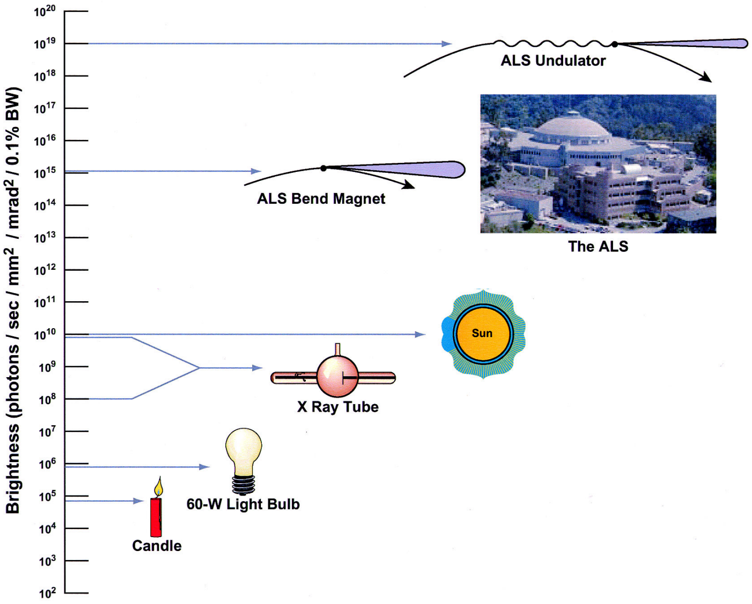


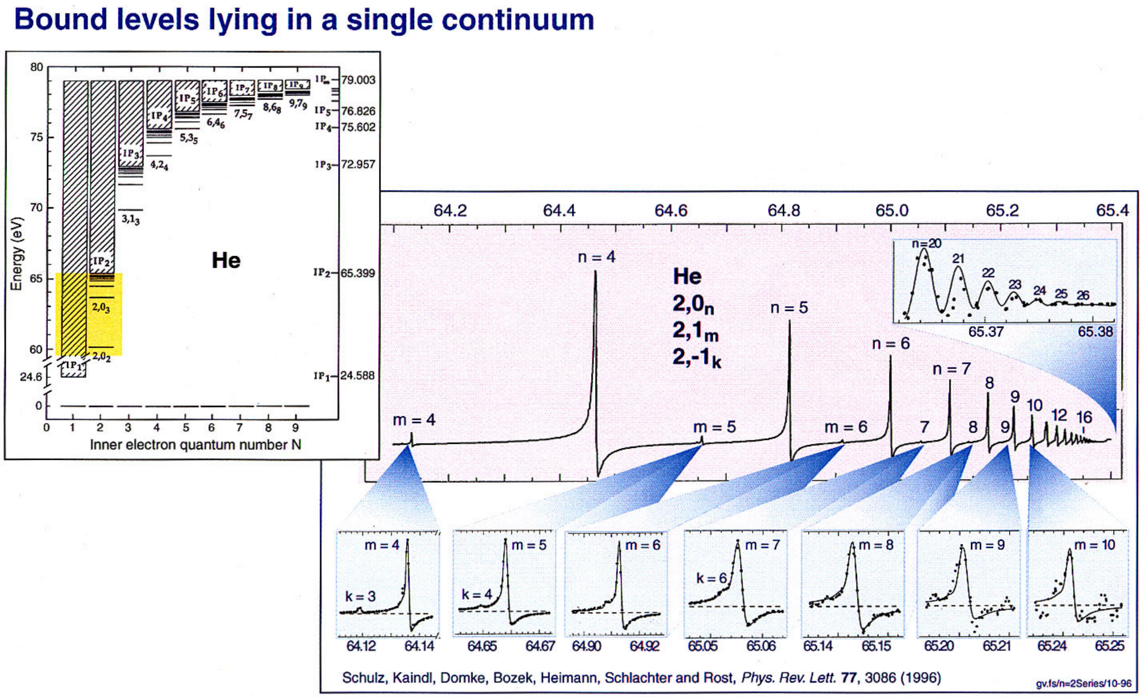
Figure 6. A photon beam from an ALS undulator is a billion times brighter than the sun. (Figure courtesy of ALS.)

Spectral resolution, photon flux, and spot size generally determine the counting rate of an experiment. The resolving power of an ALS undulator beam is typically 10,000 (10 meV at 100 eV for example) for photon energies about 30 eV. (Below 30 eV normal-incidence monochromators can be used to achieve higher spectral resolution.) However, under the best circumstances, a resolving power of 65,000 has been achieved at the ALS (and greater than 100,000 at BESSY II). This high resolving power (1 meV at 65 eV) has been essential for studies performed to search for quantum chaos.

The helium atom

The helium atom is the prototypical three-body atomic system. It has been extensively studied in absorption, electron emission, and fluorescence. The singly excited states have been known for a long time, and are found in high-school chemistry books. The doubly excited states were first studied nearly forty years ago, reported in 1963 in seminal papers by Madden and Codling [2] and Cooper et al [3]. Interference between excited levels and ionization continua caused the well-known Fano lineshape in the absorption spectrum of doubly excited states.

Little progress was made in the ensuing 30 years, until new developments in high-resolution monochromators for the soft-x-ray region of the spectrum and in high-brightness storage rings led to vastly improved absorption spectra. A plane-grating monochromator (SX-700) at BESSY was used by Domke et al [4] to achieve a resolving power of 16,000. They found the until-then-unseen third series (the 2p3d state is the first member of the series) and mapped out a considerable region of the doubly excited spectrum. The next development was the work of a Berlin/Berkeley group at the Advanced Light Source in Berkeley, where use of the third-generation high-brightness ALS light source and a spherical-grating monochromator allowed measurements with a resolving power of 65,000 [5]. These results were reported in 1996. New Rydberg series and new resonances were reported, including observation of 4 of the 5 series converging to the third ionization threshold. A very narrow resonance (2p3d in old notation, 2,-1_3 in new notation) allows measurement of the monochromator function without deconvolution. Even higher resolving power has been reported at BESSY II.



Schulz, Kaindl, Domke, Bozek, Heimann, Schlachter and Rost, *Phys. Rev. Lett.* **77**, 3086 (1996)

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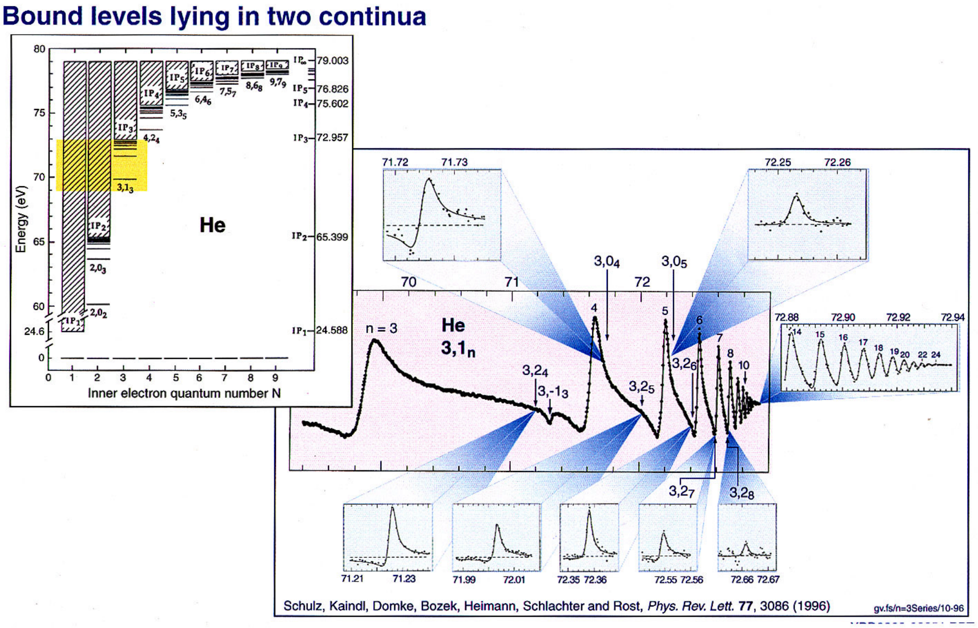
Figure 7. Absorption spectrum of helium showing states below the N=2 ionization limit [5]. Bound levels overlap a single ionization continuum, producing the three resonance series shown. The shaded area in the energy-level diagram shows the spectral region of interest.



Schulz, Kaindl, Domke, Bozek, Heimann, Schlachter and Rost, *Phys. Rev. Lett.* **77**, 3086 (1996)

Figure 8. Absorption spectrum of helium showing states below the N=3 ionization limit [5]. Bound levels overlap two ionization continua, producing 5 series of interferences (of which 4 are observed). The shaded area in the energy-level diagram shows the spectral region of interest.

The helium atom has a complex spectrum because of interference between levels and ionization continua, as reported by Madden and Codling. It has an even more complicated spectrum due to the overlap of levels which converge to different ionization levels, the so-called interlopers or perturbers.

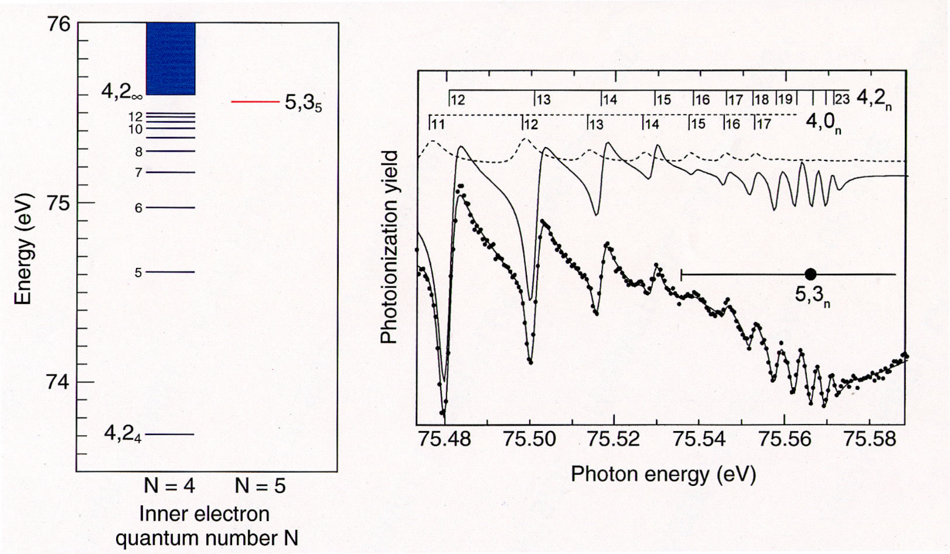


Figure 9. Absorption spectrum of helium showing states below the N=4 ionization limit [6]. The effect of the perturber $5,3_5$ state is clear.

The H$^-$ ion: a similar case

A similar three-body atomic system, the negative ion of hydrogen, has been studied extensively by Bryant et al [7].

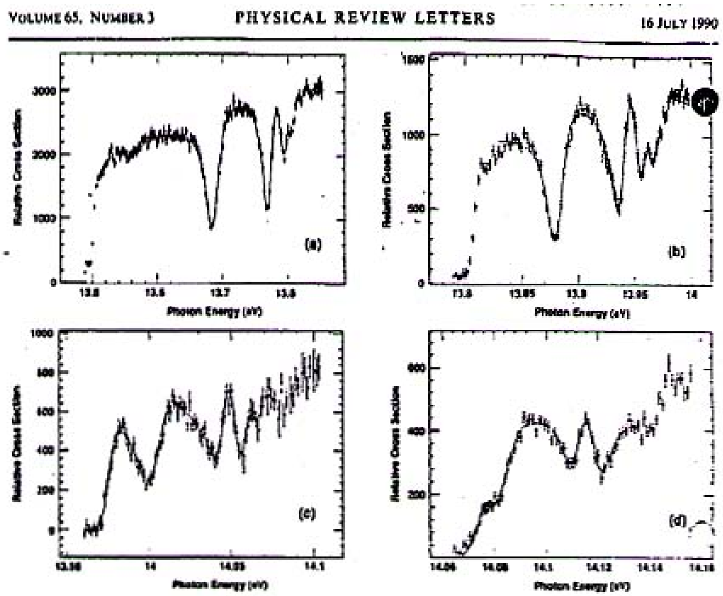


Figure 10. Partial photodetachment cross sections of H$^-$ showing production of neutral hydrogen atoms [7].

The search for quantum chaos

The information contained in this section of the paper is attributed to R. Püttner, B. Gremaud, D. Delande, M. Domke, M. Martins, A. S. Schlachter, and G. Kaindl [8].

The classical three-body problem in celestial dynamics has been known to be chaotic for at least one hundred years. By 1904 more than 800 papers had been published on the topic [9].

The classical three-body Coulomb problem should depict the same behavior as the three-body gravitational problem, because the force between charged particles is described (within a sign) by the same inverse square law with distance. Thus the helium atom is a sub-nanoscale laboratory well suited to study the three-body Coulomb problem, but from a quantum-mechanical perspective.

A fundamental physics question is how underlying classical chaos manifests in a quantum system. How does a quantum system “know” whether or not its underlying classical counterpart has chaotic behavior? We can expect to see some different quantum behavior in an atom depending on whether or not the motion of its classical counterpart is chaotic.

Classical dynamics, and thus chaos, should manifest when the atom is in a nearly classical state, or one in which both electrons are excited far from the nucleus. As a consequence, the study of high-lying doubly excited states of helium should allow the observation of a transition to chaotic behavior.

The helium atom is well described by a set of quantum numbers for singly excited and for previously studied doubly excited states. This is to be expected for a quantum system, as the Schroedinger equation is linear, and thus chaos is not admitted.

The spectrum of the helium atom shows considerable complexity close to the threshold for double ionization, i.e., when both electrons are excited far from the nucleus [6,8].

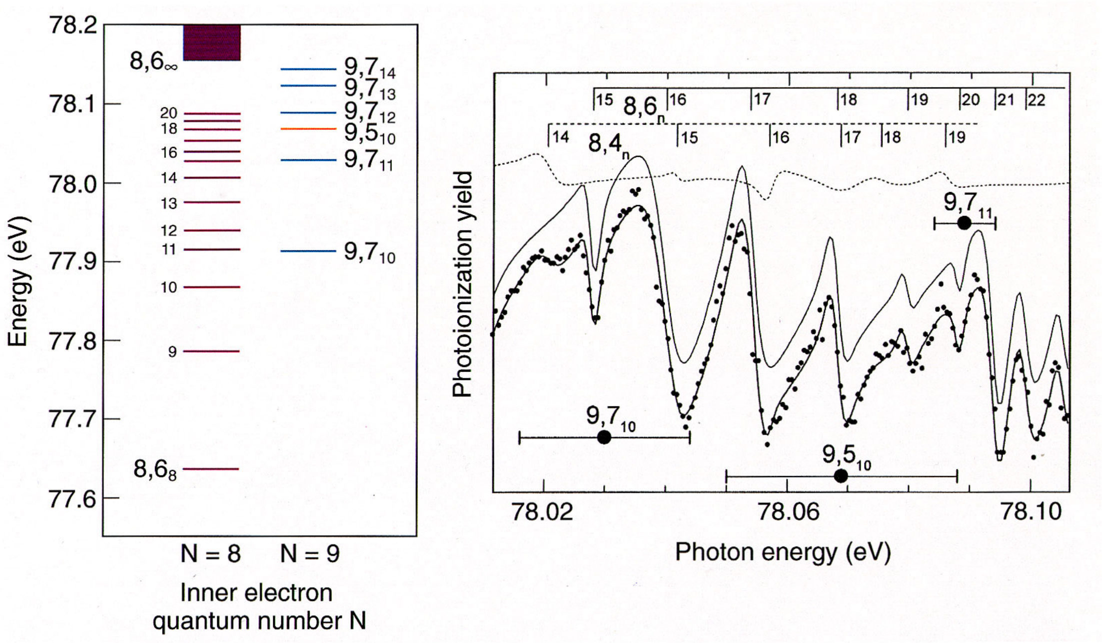


Figure 11. Absorption spectrum of helium showing states below the N=8 ionization limit [6]. There are multiple perturbing levels overlapping multiple ionization continua.

Finally, the spectrum becomes yet more complex because of the possibility of the emergence of quantum chaos.

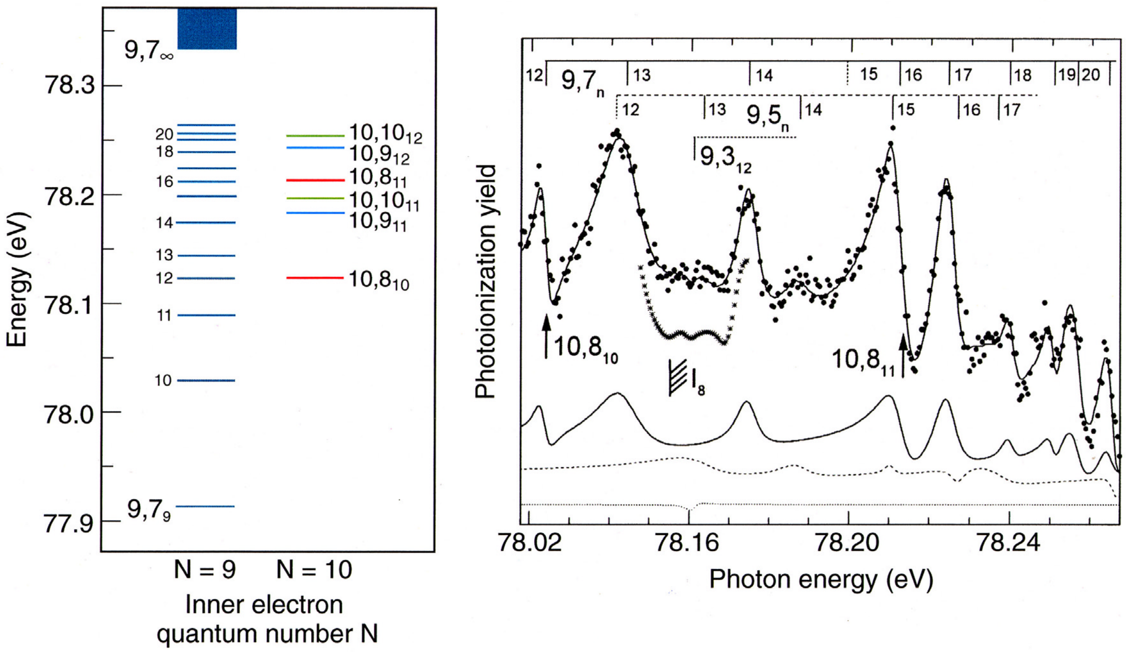


Figure 12. Absorption spectrum of helium showing states below the N=9 ionization limit [8]. The $9,7_n$ Rydberg series with the perturbers $10,8_{10}$ and $10,8_{11}$ are shown. The solid line through the data is the best fit. The solid, dashed, and dotted lines represent the $9,7_n$, 9, 5_n, and $9,3_n$ Rydberg series.

The presence or absence of chaos is determined by studying the statistics of nearest-neighbor spacing. For a regular system the spectrum is composed of essentially uncorrelated energy levels, giving rise to a Poisson distribution. For a chaotic system the distribution will have a Wigner distribution.

Analysis of experimental results and *ab initio* calculations shows for the first time that the statistical properties of the energy levels of the helium atom display a transition towards quantum chaos [8].

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