# Coherent states for Rydberg atoms

R. Scheibner
*Department of Physics and Astronomy,*
*University of New Mexico, Albuquerque 87131, USA*

(Dated: 9 December 1999)

## Abstract

This paper reviews generalized coherent states for the problem of a Coulomb potential. They allow us to describe the correspondence between the quantum mechanical expectation values and the corresponding classical variables. Within this context, the dynamics of highly excited electrons, so called Rydberg electrons, is discussed. Furthermore, the creation, observation, and stabilization of Rydberg electron wave packets is described. In particular, the preparation of an atomic electron in a state closely analogous to a Schrödinger cat state is depicted.

PACS number(s): 03.65.Ca, 32.80.Rm

# I. INTRODUCTION

Since E. Schrödinger introduced coherent states for the harmonic oscillator to obtain a quantum description of the behavior of a classical harmonic oscillator [1], it has been a challenge to generalize this idea of quantum-classical correspondence. During the last two decades, there has been progress in the development of coherent states for the bound state part of the Coulomb potential. In 1990 su(2) generalized coherent states were invented [7] and in 1996 Klauder proposed coherent states for the hydrogen atom [2]. Proposals for new descriptions of coherent states have been made for Gaussian wave packets which successfully accounted for pump-probe experiments [3, 5]. Besides the development of a theoretical representation, interesting experimental work was also done. One example is the preparation of a superposition of two atomic coherent states. Because of the immense computational effort that is required for a complete quantum treatment, classical mechanics is often a practical and also successful way to study such systems. Usually, more complex atoms, like alkali atoms, are used for experimental studies, but their high energy states can in good approximation be treated as in the hydrogen atom.

The paper will be organized in the following way. A short review on generalized coherent states according to [2, 3] will be given at the beginning. After an introduction to coherent states for the hydrogen problem, a brief discussion about their properties will follow. The paper continues with a section about creation, stabilization, and measurement of the propagation of Rydberg electrons. Then an experiment with coherently superimposed Rydberg wave packets is described and finally a conclusion is given.

# II. THEORY

## A Generalized coherent states

In the following coherent states according to the approach of Klauder are discussed. Another discription, which will not be treated in this paper, is given by Perelomov [16].

In analogy to the coherent states of the harmonic oscillator (here in polar coordinates, $\alpha = re^{i\phi}$)

$$|r,\phi\rangle \;=\; e^{-\frac{1}{2}|r|^2}\sum_{n=0}^{\infty}\frac{r^n}{\sqrt{n!}}\,e^{in\phi}|n\rangle, \tag{1}$$

where $[a,a^\dagger]=1$ and $|n\rangle$ denote normalized eigenstates of the harmonic oscillator Hamiltonian $H_0 \;=\; \hbar\omega a^\dagger a$, Klauder's coherent states for Hamiltonians with discrete spectra

$$H|n\rangle \;=\; E_n|n\rangle \;=\; \hbar\omega e_n|n\rangle \tag{2}$$

are defined by

$$|n_0,\phi_0\rangle \;=\; (N(n_0))^{-1/2}\sum_{n=0}^{\infty}\frac{n_0^{n/2}}{\sqrt{\varrho_n}}\,e^{ie_n\phi_0}|n\rangle. \tag{3}$$

Here the range of the azimuthal angle $\phi_0$ is extended from $-\infty$ to $+\infty$ which is important for the resolution of the identity operator and an essential step in Klauder's construction. $e_n$ is dimensionless and ordered by magnitude ($e_0 < e_1 < e_2 < ...$). For the harmonic oscillator $e_n \;=\; n+\frac{1}{2}$ and for the hydrogen atom $e_n \;=\; \frac{1}{(n+1)^2}$. The Klauder generalized states are usually not minimal uncertainty states, but ensure temporal stability according to Klauder's definition [2].

For the hydrogen problem, generalized angular momentum coherent states (also called su(2) generalized coherent states) are needed. According to [3] they have the form

$$|j,\Theta,\phi\rangle \;=\; \sum_{p=0}^{2j}\frac{e^{ip\phi}}{p!}\cos^{2j-p}(\Theta)\sin^{p}(\Theta)\left(\frac{(2j)!p!}{(2j-p)!}\right)^{1/2}|j,j-p\rangle, \tag{4}$$

with $|j,m\rangle$ as an eigenstate of $J^2$ and $J_z$ for the su(2) algebra of angular momentum operators. These states are normalized and provide a resolution for the identity operator.

## B Rydberg coherent states and their properties

According to Bohr's correspondence principle, atomic electrons in highly excited states should behave like classical, charged particles. That means the electron should move along

a Kepler orbit around the nucleus and stay well localized. This picture is similar to the motion of celestial bodies. In quantum mechanics it means that the expectation values of the corresponding classical variables follow the classical equations of motion and the variances of these variables do not increase with time.

Atoms in highly excited states can be described by a Hamiltonian similar to the hydrogen atom

$$H = \frac{p^2}{2m_0} - \frac{Ze^2}{r}, \tag{5}$$

which has the energy eigenvalues

$$E = -\frac{(Ze^2)^2 m_0}{2\hbar^2 n^2}. \tag{6}$$

Following Klauder's construction, Majumdar and Sharatchandra [5] and Fox [3] proposed coherent states for the hydrogen atom. With these Rydberg states it is possible to describe the dynamics of the electron as characterized by the expectation values of the angular momentum $\vec{L}$ and the Runge-Lenz vector (eccentricity vector) $\vec{\epsilon}$ both defined by

$$\vec{L} = \vec{r} \times \vec{p} \qquad and \qquad \vec{\epsilon} = \vec{r} - \frac{1}{2Ze^2 m_0}(\vec{p} \times \vec{L} - \vec{L} \times \vec{p}). \tag{7}$$

Under the assumption of conserved angular momentum and eccentricity, Fox shows that the expectation values of $\vec{L}$ and $\vec{\epsilon}$ follow the classical equations of motion and that the variances of these two operators are time independent. Also the ratio of the square root of the variance and the principle quantum number $n_0$ decreases with increasing $n_0$. Furthermore it can be shown, depending on the chosen state, that the expected value of the position follows a circular or slightly eccentric orbital motion. Nevertheless, it is also important to know the development of the uncertainty (variance) in the coordinates corresponding to the classical variables r, $\Theta$, and $\phi$. By knowing the explicit coordinate dependence of the Rydberg states ($\psi_{Ryd}(r, \Theta, \phi, t) = \langle r, \Theta, \phi | Ryd, n_0, \phi_0, t \rangle$) it is possible to calculate these values. For circular Rydberg states the root-mean-square deviation divided by the mean is

$$\frac{\sqrt{\langle \Delta^2 \Theta \rangle}}{\pi/2} = \frac{\sqrt{2}}{\pi\sqrt{n_0}} \qquad for\ \Theta \tag{8}$$

and

$$\frac{\sqrt{\langle \Delta^2 r \rangle}}{r_0} = \frac{1}{\sqrt{2n_0}} \qquad for\ r. \tag{9}$$

Hence, for sufficiently large principle quantum numbers $n_0$, $\Theta$ is localized in the azimuthal plane and r is relatively close to the circle radius. This is in agreement with the quantum-classical correspondence discussed at the beginning of this section. In contrast to these time independent results, the distribution for the azimuthal angle $\phi$ is time dependent and the variance increases within a Kepler period by a factor of $\approx 10^3$ for typical experimental Rydberg atom states with $50 \leq n_0 \leq 200$. That means that there will be a total dephasing in $\phi$ after less than one orbital period. Fox points out that the dephasing at least improves when celestial mechanics is treated by this approach. That means for $n_0 \approx 10^{74}$ in the case of the system sun and earth. By this, he demonstrates the quantum-classical correspondence principle for very large quantum numbers.

The rapid dephasing in the azimuthal angle for these coherent states has also been demonstrated by Bellomo and Stroud [4] by using the time autocorrelation function proposed by Nauenberg [11]

$$C(t) = \left| \langle \psi | exp \left[ -\frac{i}{\hbar} H t \right] | \psi \rangle \right|^2 \tag{10}$$

(in a.u.) with $|\psi\rangle$ standing for a generalized coherent state or a wave packet. The autocorrelation function contains information about when and how a state returns to its original configuration. When representing an electron on a Kepler orbit, it should return to one or very close to one, which is the maximum value for normalized states. Figure 1 shows the result of the time evolution of the autocorrelation function for principle quantum numbers $n_0 = 25$ and $n_0 = 400$.

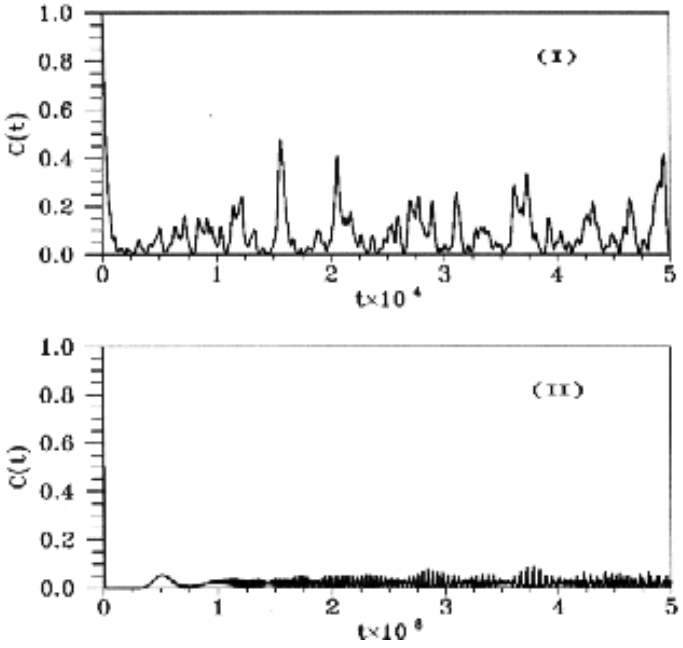


FIG. 1: Autocorrelation for temporarily stable states: in (a) $n_0 \approx 25$, in (b) $n_0 \approx 400$. The small recurrence peaks of (a) disappear in (b).

## III. EXPERIMENTS

### A Creation and measurement of Rydberg atoms

As in the experiment described below, very often other elements, e.g. alkali metals, than hydrogen are used to produce Rydberg atoms, since it is much easier to excite their valence electron to a Rydberg state. The excitation energy is in the UV spectrum of the light and the pulse durations for a coherent excitation are on a femto- to picosecond scale. The experimental surrounding conditions have to be chosen carefully, since thermal collisions can ionize the highly excited atoms. The binding energy of hydrogen with, for example, $n_0 = 100$ is only $1.36 \times 10^{-3}\, eV$ and the energy separation between two energy eigenstates $\Delta E$ is of the magnitude $10^{-5}\, eV$ [12]. The wave packet is composed of the eigenstates spanned by the frequency bandwidth of the laser pulse around the mean excitation level $n_0$ and oscillates between the nucleus and the classical outer turning point at $r_C = 2n_0^2$ (in a.u.). To measure the quantum state distribution of the excited electron, state-selective field

ionization is used. Here, a DC electric field is ramped on. It ionizes the population from different excitation levels at different times. The produced ions are collected with an electron multiplier and counted according to their arrival time. The resulting time-resolved ion signal has peaks which correspond directly to different Rydberg eigenstates in the superposition. By varying pulse length and field distribution, it is possible to create more complicated wave-packet structures than the ones with a Gaussian shape. To obtain such wave packets, the pulse can be considered as a superposition of electric fields that would create separate substructures.

Creating simple Rydberg packets or even shaped wave packets is only the first step towards demonstrating the control of electronic dynamics in packets. According to reference [9], a radially localized Rydberg electronic wave packet can be formed by a short light pulse, creating a coherent superposition of Rydberg states with an average principle quantum number $n_0$. The corresponding classical system of this wave packet is an ensemble of classical orbits with arbitrary orientations in space around the nucleus but with synchronized phase of the electron in each classical realization. Therefore, the entire ensemble appears to be a shell oscillating in and out from the nucleus. After the excitation pulse the wave packet is allowed to freely evolve on its elliptical orbit for a delay time $\tau_d$. Then the state of the atom is probed by a second short optical pulse. By analyzing the photo-ionization signal as a function of the delay time, the evolution of the wave packet can be observed since the ionization probability is larger when the wave packet is near the nucleus.

The interaction of the pulse with the wave packet can be understood classically. When the electron is near the nucleus it moves faster than when it is near the outer turning point. The rate of absorption of the second pulse is given by the product of the electron current $\vec{J}$ which is proportional to its velocity and the electric field of the pulse. Therefore, the photo ionization will be lower when the electron is near an outer turning point.

Since the energy levels of the hydrogen atom are neither equally (harmonic oscillator) nor randomly spaced, the superposition of the states, represented by the wave packet, neither stays unchanged, as for the harmonic oscillator coherent states, nor does it spread and

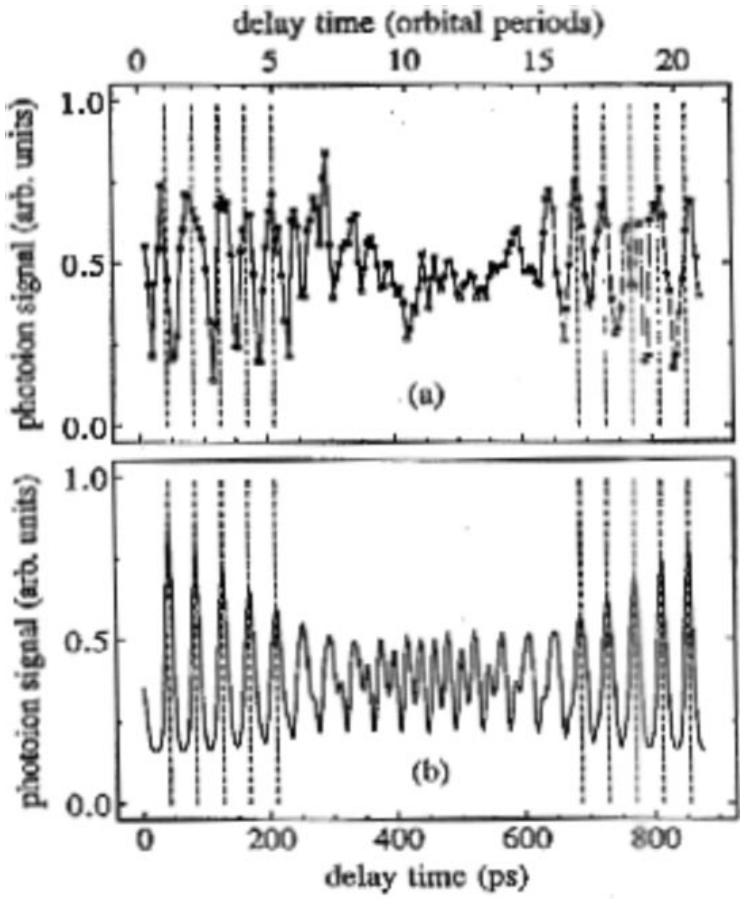


FIG. 2: Photo-ionization signal as a function of the delay time: (a) measurement, (b) theoretical prediction. A classic-non classic-classic evolution cycle with a half fractional revival is shown.

never revive. The temporal measurement shows a fast oscillation representing the electron wave packet on its Kepler orbit which decays and revives after some time with a periodicity proportional to the second derivative of the average wave packet energy. Between the periods of time when the wave packet can be described classically it is possible to observe fractional revivals. A half fractional revival, for example, means that the electron relocalizes in two separated wave packets and the frequency of the photo ionization signal doubles. The revivals of the wave packets are not complete, because there is always a coupling of the system to sources of decoherence as there is for example the blackbody radiation. An example of a comparison between theoretical expectation and experimental results of [9] is shown in Fig.2.

## B Stabilization of Rydberg wave packets

Ultra-stable Rydberg states are those with maximum angular momentum, since the interaction between electron and nucleus is then minimal. However, the excitation of the electron with one pulse usually creates states with low angular momentum. To avoid core effects, which enhance the decay rate of the excited states, it is necessary to increase the angular momentum of the electron. This is achieved by slowly varying, weak electric, and magnetic fields [4, 10]. In order to assemble a wave packet with little or no dispersion in all variables, a quantum system with (almost) constant energy-level spacings is necessary as in the case of the harmonic oscillator. This can be achieved by weak external fields. Lee *et al.* [6] show that it is possible to create global equilibrium points with circularly polarized microwave and magnetic fields (in a rotating frame). For non-dispersing wave packets these equilibrium points have to be stable by themselves and the potential of the surrounding area has to be harmonic in a region that is large compared to the wavelength of the electron. Otherwise there would be dispersion out of the region caused by tunneling. The states obtained under these conditions are approximately the conventional coherent states, since the local Hamiltonian is harmonic. Lee *et al.* point out, that the equilibrium points are analogous to those in the restricted three- body problem in celestial mechanics. It is also possible to manipulate their properties by modifying the external fields so that the wave packets stay localized. This classical treatment of the coherent states is justified as long as the fields used are weak compared to the Coulomb field the electron senses [10].

## C Schrödinger cat state

Schrödinger brought the distinction between the descriptions of the reality in classical and quantum physics into sharp focus by introducing the quantum superposition of a “dead cat” and a “live cat”. Basically, it is a quantum mechanical superposition of two classically distinguishable physical states which are “localized” in one or the other only by the act of

a measurement.

With the preparation of a coherent superposition of two Rydberg wave packets in 1995, Noel and Stroud [13] succeeded in creating such a Schrödinger cat state of nearly macroscopic size. They formed the Schrödinger cat state of the atom by the interaction with a pair of phase-coherent time-delayed laser pulses. The time delay was half the time the electron needs for a Kepler period. This insured that the first part of the wave packet was at the outer turning point, when the second pulse excited the second wave packet at the inner turning point. The result of this sequence is a single electron in a superposition at two well separated spatial locations. The measurement of the quantum state distribution for various phase differences between the two wave packets showed that the presence of a second wave packet modifies the state distribution compared to that of a single wave packet. It is possible, for example, to create even or odd coherent states. They are composed of only even or odd eigenstates $|n\rangle$. This modification of the population distribution is not the only indication of the interference between the two quasi-classical wave packets. The characteristic interference between them in phase space can also be observed for the Schrödinger cat state.

However, this observation can only be made when the two wave packets spatially overlap and are near to the core. Since the energy spacing between the eigenstates is not constant the initially, spatially separated wave packets spread and overlap sufficiently after some time. To measure the interference between the wave packets a time-delayed probe pulse identical to the excitation pulse is sent to the atom to determine the location of the radial electron wave packet. This probe pulse interacts coherently and a rapid oscillation in the excited state population at the optical period can be seen. The modification of the population distribution and the oscillation of the fringe visibility with changing phase between the two wave packets in Noel's and Stroud's experiments showed that the classically distinguishable wave packets are created in a coherent superposition (Schrödinger cat state).Unfortunately, the size of such Schrödinger cat states is limited by the time scale for mixing due to blackbody transitions which goes as $n^2$. Since the period of the Kepler period scales with $n^3$ there is a point at which the coherence between the highly excited wave packets will be lost before

they can complete even a single orbit.

## IV. CONCLUSION

By the description of Rydberg coherent states, a manifestation of the quantum-classical correspondence principle at high quantum numbers has been made. The behavior of the proposed states is almost in complete agreement with the measurements. Nevertheless, the problem of the azimuthal dephasing has not been solved satisfactorily so far. Besides this correspondence between quantum mechanical expectation values and classical variables, the Rydberg-atom system still maintains pure quantum mechanical properties. The experiments done with Rydberg atoms show us that there are other properties in addition to the size of a system which distinguish the classical limit from the quantum case. The dependence of the evolution of the wave packets on the initial excitation allows us to manipulate the transitions of electrons in many ways and thus control the outcome of material processes [15]. In this respect, the increase of the efficiency of chemical reactions would be an application of interest. Besides these practical applications, the variety of semi-classical and quantum features has made and will continue to make Rydberg atoms object of future studies.

## REFERENCES

[1] E. Schrödinger, Naturwissenschaften **14**, 644 (1926).

[2] J.R. Klauder, J. Phys. A **29**, L293 (1996).

[3] R.F. Fox, Phys. Rev. A **59**, 3241 (1998).

[4] P. Bellomo, and C.R. Stroud, Jr., J. Phys. A. **31**, L445 (1998); P. Bellomo, and C.R.

Stroud, Jr., Phys. Rev. A **59**, 900 (1998).

[5] P. Majumdar, and H.S. Sharatchandra, Phys. Rev. **56**, 3322 (1997).

[6] E. Lee, A.F. Brunello, and D. Farrelly, Phys. Rev. A **55**, 2203 (1996).

[7] R.F. Fox, and T.C. Elston, Phys. Rev. E **47**, 282 (1993).

[8] P. Bellomo, and C.R. Stroud, Jr., Phys. Rev. **59**, 2139 (1998).

[9] J.A. Yeazell, M. Mallalieu, and J.R. Stroud, Jr., Phys. Rev Let. **64**, 2007 (1990).

[10] P. Bellomo, C.R. Stroud, Jr., D. Farrelly, and T. Uzer, Phys. Rev. A **58**, 3896 (1998).

[11] M. Nauenberg, *Coherent States: Past, Present and Future* ed D. Feng *et al.* (Singapore, World Scientific, 1994).

[12] B.H. Bransden, and C.J. Joachain, *Physics of Atoms and Molecules* (Longman Scientific & Technical, New York, 1995).

[13] M.W. Noel, and C.R. Stroud, Jr., Phys. Rev. Lett. **77**, 1913 (1996); M.W. Noel, and C.R. Stroud, Jr., Phys. Rev. Lett. **75**, 1252 (1995).

[14] J. Parker, and C.R. Stroud, Jr., Phys. Rev. Lett. **56**, 716 (1986).

[15] W.S. Warren, H. Rabitz, and M. Dahleh, Science **259**, 1581 (1993); P. Brumer, and M. Shapiro, Annu. Rev. Phys. Chem. **43**, 257 (1992); D.J. Tannor, and S.A. Rice, Adv. Chem. Phys. **70**, 441 (1988).

[16] A. Perelomov *Generalized Coherent States and their Application* (Springer-Verlag, Berlin, New York, 1986).