

Measurement of the Amplitude and Phase of a Sculpted Rydberg Wave Packet

T. C. Weinacht, J. Ahn, and P. H. Bucksbaum

Physics Department, University of Michigan, Ann Arbor, Michigan 48109-1120

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We have completely determined the amplitude and phase of a quantum wave packet. The wave packet was shaped using a programmable optical pulse. Phase information comes from analysis of covariant fluctuations due to interference between the wave packet and a well-characterized reference. [S0031-9007(98)06267-X]

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We have measured the amplitude and quantum phase of sculpted Rydberg wave packets in atomic cesium, by adapting techniques used for measuring the classical phase of a light pulse.

The stationary Rydberg states ψ_{nlm} are real functions of θ and r up to an unobservable global phase factor $e^{i\delta}$. Here (n, l, m) are the usual quantum numbers specifying the energy of the state, its angular momentum, and z component. The azimuthal dependence of ψ_{nlm} is a complex exponential, $e^{im\phi}$, which will not concern us here.

Nonstationary states, called wave packets, are not real functions of θ and r . They are coherently phased superpositions of nondegenerate eigenstates. Figures 1(a) and 1(b) show an example: a radial Rydberg wave packet made up of eight np states in atomic cesium, observed near the outer-turning point [1]. This figure shows a cross section in the x - z plane. The phase is represented by color. The upper picture shows the measurement of a sculpted wave packet; below it is the calculated model wave packet. Now we will explain how this wave packet was made and measured.

The key to determining Ψ is decomposition into its constituent eigenstates, $\psi_i(r, \theta, \phi)$:

$$\Psi(\mathbf{r}, t) = \sum_i a_i \psi_i(r, \theta, \phi) e^{-i\omega_i t}. \quad (1)$$

If the eigenstates and eigenvalues are known or can be measured, then the wave packet is completely specified by measuring the amplitude and phase of the complex coefficients a_i . For example, the superposition of eight eigenstates in Fig. 1(b) has real a_i 's that alternate in sign. Table I shows the measured phases and amplitudes for Fig. 1(a). This phase information was never available using previous detection methods for the wave packet.

The amplitude of the a_i 's can be analyzed by state-selective field ionization (SSFI) [2], since each nondegenerate eigenstate in the wave packet has a unique field-ionization threshold. A second approach is wave packet interferometry, also called the optical Ramsey method [3]. Here two wave packets are summed coherently by exciting the atom with two identical light pulses with a variable time-delay τ . If the usual weak-field condition for time-dependent perturbation theory applies, then the wave function is given by

$$\Psi(\mathbf{r}, t, \tau) = \sum_i (a_i \psi_i e^{-i\omega_i t} + a_i \psi_i e^{-i\omega_i(t+\tau)} e^{i\omega_{gs}\tau}), \quad (2)$$

where $e^{i\omega_{gs}\tau}$ represents the phase advance of the initial state during the delay time τ . The excited state population $\langle \Psi(\mathbf{r}, t, \tau) | \Psi(\mathbf{r}, t, \tau) \rangle$ oscillates with the time delay τ :

$$\langle \Psi(\tau) | \Psi(\tau) \rangle = \sum_i 2|a_i|^2 [1 + \cos(\omega_{gs} - \omega_i)\tau]. \quad (3)$$

The spectral amplitude can be extracted by Fourier analysis of this interference function, but the phase information is missing; therefore the shape of the wave packet cannot be determined.

The phase problem in Rydberg wave packets has an analog in ultrafast optics. There are several techniques to determine the spectrum and phase of the electric field in a subpicosecond laser pulse [4]. A recent paper suggested that *spectrally resolved cross correlation*, the basic method used in spectral interferometry [5], was also directly applicable to the phase problem in wave packets [6]. This suggestion was the starting point for the work reported here.

Cross correlation is similar to optical interferometry, where the signal received by a detector is the coherent sum of two optical fields, $E(t)$ and $E'(t)$, combined on a beam splitter. In conventional Michelson interferometry the two fields are the same except for a time-delay τ , and so the detector sees an autocorrelation of the electric field superimposed on a background:

$$\begin{aligned} S_{\text{auto}}(\tau) &= \int |E(t) + E(t + \tau)|^2 dt \\ &= 2 \int E(t)E(t + \tau) dt + 2 \int |E(t)|^2 dt. \end{aligned} \quad (4)$$

Note that all phase information is lost, and that S_{auto} has a similar formal appearance to the wave packet signal in Eq. (3). In a cross-correlation, the second pulse is replaced by a reference field whose properties are known,

$$\begin{aligned} S_{\text{cross}}(\tau) &= 2 \int E(t)E_{\text{ref}}(t + \tau) dt \\ &\quad + \int [|E(t)|^2 + |E_{\text{ref}}(t)|^2] dt. \end{aligned} \quad (5)$$

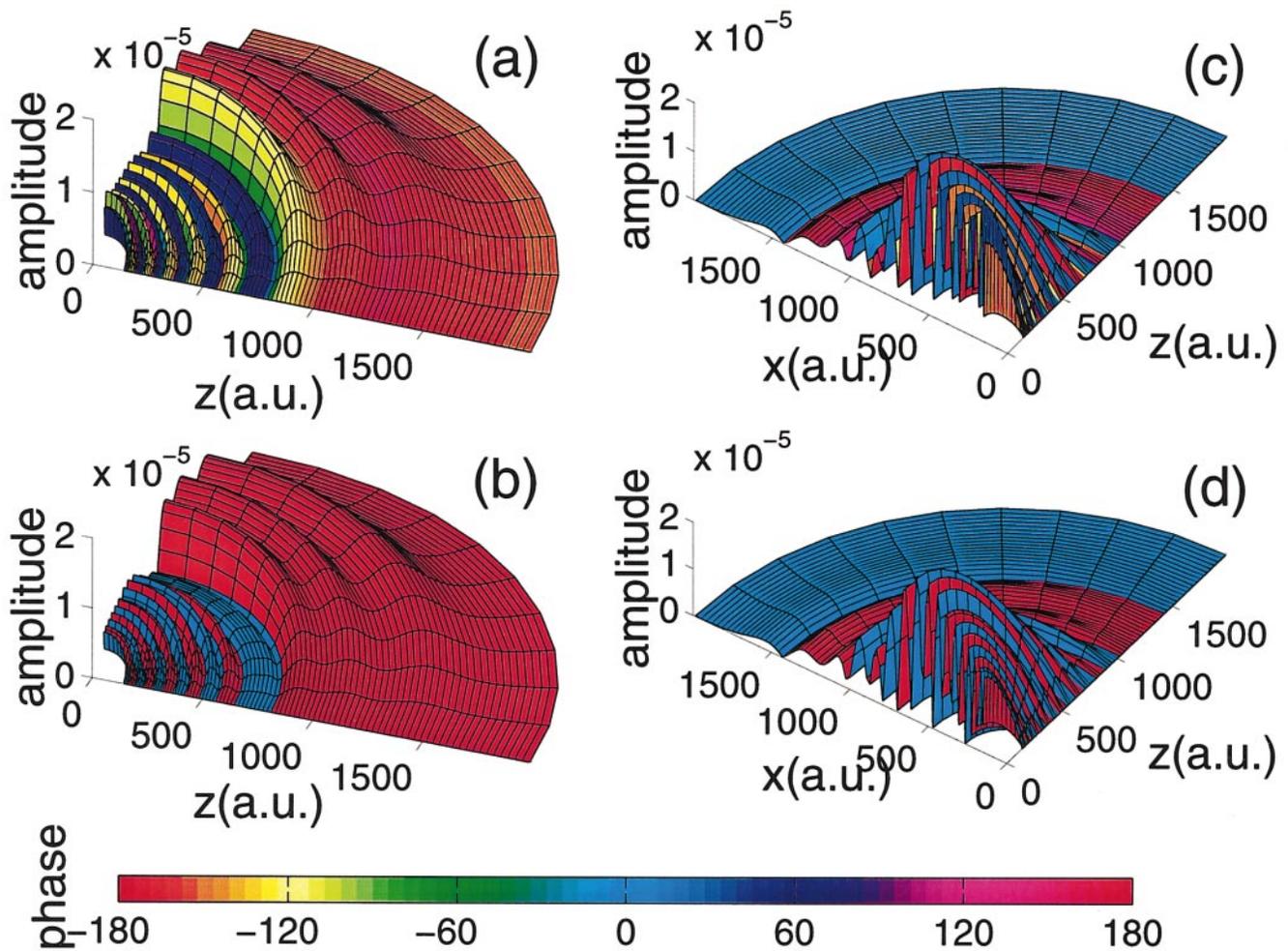


FIG 1(color). (a) Measured Rydberg wave packet amplitude and phase, for the eight cesium np states coherently superposed as described in Table I for wave packet 1. Phase is described by color, and amplitude by height. Only one quadrant of the x - z plane is shown. (b) Calculated wave packet with the same eight states as (a). Each state has equal amplitude and alternating sign. (c) Measured Rydberg wave packet amplitude and phase of wave packet 2 described in Table I for wave packet 2. (d) Calculated wave packet with the same eight states as figure (c). Each state has equal amplitude. The lower four states have $\delta_{ij} = 0$; the upper four states have $\delta_{ij} = \pi$.

The first term is then a convolution integral, which can be Fourier decomposed to reveal the phase difference between each frequency component of E and E_{ref} . The phase of the reference light pulse can be analyzed using methods such as frequency-resolved optical gating (FROG) [4]. Spectral interferometry is a way to extend the measurement of optical phase to pulses that do not lend themselves easily to FROG analysis.

When “reference” and “signal” quantum wave packets interfere, the phase difference between each pair of eigenstates in the signal can be resolved through the temporal interference pattern of each state in the spectrum. Thus, spectral interferometry can be applied in quantum mechanics.

Our measurements were performed on an effusive beam of cesium atoms, which were prepared in the $7s$ state by a two photon transition from the $6s$ ground state via a

10 ns, $1.08 \mu\text{m}$ pulse from a Raman-shifted dye laser. The signal and reference wave packets were produced by excitation from the $7s$ state to the Rydberg np series, using the output of a chirped-pulse amplifier system. A Kerr-lens mode-locked titanium sapphire laser, followed by a dispersive grating-pulse expander was used to seed a 10 Hz regenerative amplifier. Part of the light was directed into an optical pulse shaper, which consisted of a spectrally resolved acousto-optic Bragg deflector (AOM) at the midpoint of zero-dispersion stretcher [7]. The amplitude and phase of the acoustic waveform in the AOM was programmable, so the optical pulse could be reshaped to any field consistent with the available bandwidth. The shaped laser pulse was further amplified in a Ti:Sapphire cell, and then recompressed in a parallel grating compressor. This light created a Rydberg wave packet Ψ_{signal} at time $t = 0$:

TABLE I. Measured amplitude and phase in degrees for each eigenstate in wave packets 1 (wpkt 1) [Figure 1(a) and 1(b)] and 2 (wpkt 2) [Figure 1(c) and 1(d)]. Amplitude errors are non-statistical estimates of maximum systematic deviations during data collection. Phase errors are given by the standard deviation in fitting the phase to the acquired cosine curve.

n state	Amplitude	wpkt 1 phase	wpkt 2 phase
25	0.44 ± 0.02	53.3 ± 4.0	200.5 ± 4.0
26	0.39 ± 0.02	211.0 ± 5.2	197.6 ± 5.1
27	0.37 ± 0.02	2.9 ± 10.3	208.6 ± 5.1
28	0.37 ± 0.02	188.1 ± 5.7	171.3 ± 4.5
29	0.33 ± 0.02	0.0	0.0
30	0.31 ± 0.02	163.5 ± 5.2	0.0 ± 16.0
31	0.30 ± 0.02	10.9 ± 0.6	0.6 ± 6.3
32	0.31 ± 0.02	205.3 ± 5.2	0.0 ± 5.4

$$\Psi_{\text{signal}}(t = 0) = \sum_i a_i \psi_i, \quad a_i = |a_i| e^{i\delta_i}. \quad (6)$$

The remainder of the light from the regenerative amplifier went to a second grating-pulse compressor, yielding a 150 fs pulse. This reference pulse contained a small amount of high-order dispersion; however, the phase of the spectrum over the bandwidth of the experiment was constant to within ± 0.5 radians, or about 0.05 radians between neighboring n states. This pulse excited the same cesium atoms at $t = \tau$, creating a second (reference) wave packet:

$$\Psi_{\text{ref}}(t = \tau) = e^{i\omega_{\text{gs}}\tau} \sum_i b_i \psi_i, \quad b_i \in \Re. \quad (7)$$

The resulting total wave function can be written as

$$\Psi_{\text{total}}(t = \tau) = \sum_i (a_i e^{-i\omega_i\tau} + b_i e^{i\omega_{\text{gs}}\tau}) \psi_i. \quad (8)$$

The measured quantity for each laser shot was the total population (P_i) in each Rydberg state as determined from SSFI spectra:

$$P_i = |a_i|^2 + |b_i|^2 + 2|a_i||b_i| \cos[(\omega_i - \omega_{\text{gs}})\tau - \delta_i]. \quad (9)$$

From the P_i , we wished to extract the relative phases between the states in the wave packet: $\delta_{ij} = \delta_i - \delta_j$.

P_i oscillates at the optical frequency. The time-delay τ should therefore be stable to much better than the optical period of about 2 fs, which is not practical in a system with amplifiers and pulse shapers in separate delay lines. Furthermore, the carrier frequency in the radio-frequency waveform in the AOM is not phase locked to the laser, resulting in a random global phase shift on the signal pulse even in the absence of jitter in the time delay. Therefore, over several laser shots, the cosine term in P_i averages to zero, so the average values $\langle P_i \rangle$ and the rms populations (ΔP_i) have no phase dependence:

$$\langle P_i \rangle = |a_i|^2 + |b_i|^2, \quad (10)$$

$$(\Delta P_i)^2 = 2|a_i|^2|b_i|^2. \quad (11)$$

To extract the phase despite the time-delay instability, we construct the correlation function [8],

$$r_{ij} = \frac{\langle P_i P_j \rangle - \langle P_i \rangle \langle P_j \rangle}{(\Delta P_i)(\Delta P_j)}. \quad (12)$$

Even after averaging over the optical frequency, the product $\langle P_i P_j \rangle$ still has a dependence on the phase difference $\omega_i - \omega_j$:

$$\begin{aligned} \langle P_i P_j \rangle &= (|a_i|^2 + |b_i|^2)(|a_j|^2 + |b_j|^2) \\ &\quad + 2|a_i||b_i||a_j||b_j| \cos[(\omega_i - \omega_j)\tau - \delta_{ij}]. \end{aligned} \quad (13)$$

Inserting this into the correlation function yields a simple expression for δ_{ij} ,

$$r_{ij} = \cos[(\omega_i - \omega_j)\tau - \delta_{ij}]. \quad (14)$$

The AOM was programmed to linearly increment the phase between each eigenstate n_i and a reference state n_j by an amount $\alpha = 0$ to 4π , in 40 steps. Then $r_{ij}(\delta_{ij}, \alpha)$ was fit to a cosine function to extract the initial phase δ_{ij} with greater accuracy and precision than would have been possible from a single measurement of the correlation function. The standard error in the fit for δ_{ij} is the phase uncertainty listed in Table I. The compositions of the measured wave packets in Fig. 1 are given in Table I. The principal quantum numbers were determined by measuring the field ionization threshold for cesium atoms excited by the reference pulse and comparing it to the critical field given by $F_{\text{crit}}(n) = (2E_n)^{-2}$ (atomic units). All contributing states had $l = 1$ and were polarized along x . The amplitude of the packet is fairly close to the target shape; phase errors of up to one radian have not changed the basic form of the wave function probability density.

Figures 1(c) and 1(d) show a different wave packet, in which we attempted to maintain a constant relative phase in the lower four n states and then reverse the phase in the higher four n states. Such a sudden phase reversal half-way through an optical spectrum produces a sharp intensity minimum in the center of a broad pulse. In the highly dispersive Rydberg spectrum, the situation is far more complicated, as the figure shows.

Previous measurements of the shape of Rydberg wave packets have included the following: photoionization by a short probe pulse [9]; excitation or ionization by a short electric field pulse, known as a ‘‘half-cycle pulse’’ [10]; probing by photoexcitation of a core state, followed by autoionization [11]; and the atomic streak camera [12]. Each of these methods has achieved some success in evaluating the shape of the wave packet and each has specific advantages and disadvantages. Photoionization is sensitive to $|\Psi(\mathbf{r}, t)|_{\mathbf{r}=0}^2$, i.e., the temporal oscillations of the wave packet probability density near the core. Half-cycle pulses measure the momentum distribution $|\Phi(\mathbf{p}, t)|^2$. Without information about the phase of Φ , the Fourier transformation into Ψ is not unique. The core excitation is promising but, ultimately, also has limited resolution, except near the ion core. Finally, the atomic streak camera can measure wave packet motion in the

direction of the probing electric field, but it still cannot measure the quantum phase. In fact *none of these methods has succeeded in measuring the quantum phase structure of a Rydberg wave packet.*

Quantum wave packets in systems other than Rydberg states have also been studied, and measurements of quantum phase are not new. Wave packet motion leads to oscillating multipole moments which can be observed by free-induction decay. This is the physical principle behind most quantum-beat spectroscopies and many magnetic resonance techniques. Our covariance cross-correlation measurement is not dependent on the presence of oscillating multipole fields, however, and would work even for an s -state wave packet, which has no oscillating moments.

Rovibrational wave packets in diatomic molecules have been analyzed by laser-induced fluorescence or photoexcitation to extract their shapes [13]. Also, recent work on translational wave packets of cooled atoms confined in optical lattices have succeeded in measuring the vibrational phase by analyzing optical phase shifts accompanying absorption [14].

In conclusion, we have measured the amplitude and relative phase of a Rydberg wave packet. This constitutes a complete determination of the wave function, since the global phase is not an observable. Our technique is a spectrally resolved covariance of the wave packet interference with a well-characterized reference. This captures the phase of the measured wave packet at an arbitrary target time, when the phases of the reference wave packet are all relatively real. Our technique allows us to measure a wave packet at any point in its orbit. We purposely chose test wave packets that had interesting shapes far from the ion core, and had no difficulty extracting the amplitude and phase of these wave functions. This method is the quantum analog of optical spectral interferometry.

This measurement method could be extended to any wave packet whose constituent eigenstates are nondegenerate, and whose eigenvalues are known or can be measured. Our wave packets were made with optical pulses shaped by a spectrally resolved acousto-optic modulator, but the wave packets could be made in any way that can be synchronized to a reference. Since the measurement is made by observing covariant fluctuations (COIN technique), the synchronization need only match the time scale of the motion of the wave packet itself; interferometric stability down to an optical wavelength is not necessary.

There are many potential applications and extensions of this work. A similar interference technique based on quantum beats in fluorescence was recently proposed to study wave packets in coherently excited molecules [15].

We plan to examine the effect of half-cycle pulses on Rydberg wave packets, during and after the passage of the field pulse. In addition, recent papers have suggested methods to produce gratings in Rydberg wave packets [16]. Diffraction experiments would be sensitive to phase as well as amplitude, and our technique could be used to analyze such structures. Finally, adaptive control experiments can use phase information for feedback.

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- [1] Figure 1 shows states of pure l , neglecting the spin-orbit interaction. This is justified, because our measurements take place on a time scale (2 ps) that is short compared to the inverse frequency of the spin-orbit splitting in these states of the atom (about 60 ps in our case). Therefore we are both making and probing superposition states in the atom that correspond to pure l states, not j states. The same comment can be made to justify the neglect of the hyperfine interaction and the total angular momentum $\mathbf{F} = \mathbf{I} + \mathbf{J}$.
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