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Measurement of the quantum state of electronic wave packets

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A technique for determining the quantum state of electronic wave packets from probability distribution measurements is presented. Specifically, a measurement of the fractional probability in each of a wave packet's constituent eigenstates can be used in conjunction with a measurement of the wave packet's time-dependent probability distribution to recover the complex amplitude of each constituent eigenstate. The method is demonstrated for radial Rydberg wave packets using previously measured time-dependent probability distributions in coordinate and momentum space. By exploiting delay imaging techniques, it should be possible to retrieve the quantum state of wave packets in real time.

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I. INTRODUCTION

The control of quantum dynamics in atomic systems, both in the internal and external degrees of freedom, continues to be an important theme in atomic physics research. In the case of internal dynamics, the development of methods to monitor the coherent evolution of electronic wave packets has proven to be a real challenge [1]. Nevertheless, the ability to quantitatively assess the quantum state of a system at a specific time is crucial to many applications of quantum control [2,3].

A number of different probe methods for studying the evolution of Rydberg wave packets have been developed including short-pulse photoionization [4], bound-state interferometry [5–8], time-resolved isolated core excitation (TRICE) [9], impulsive momentum retrieval (IMR) [10,11], and the atomic streak camera [12]. These methods allow measurement of the wave packet's probability distribution near the ion core, the degree to which two wave packets resemble each other, the radial probability distribution, the momentum-space probability distribution, or (for continuum wave packets) the electron flux leaving the atom, respectively. In the absence of additional information, none of these methods allows one to determine the full time-dependent electronic wave function.

However, it was recently shown theoretically [13] and experimentally [14] that a wave-packet cross-correlation measurement combined with complete information regarding the distribution of probability among the wave packet's constituent eigenstates [15] could be used to extract the complex eigenstate amplitudes in the packet. Because the eigenenergies and wave functions for electronic Rydberg states are very accurately known, the determination of the complex eigenstate amplitudes is equivalent to a measurement of the time-dependent wave function or quantum state.

There are, however, a number of technical problems which make complex amplitude retrieval using the cross-correlation method difficult for arbitrary wave packets. Cross correlation requires the creation of two wave packets: a reference packet whose quantum state is known, and a target packet whose eigenstate distribution is known [13]. For many types of wave packets, eigenstate composition is easily measured using state-selective field ionization (SSFI). For example, radial wave packets composed of eigenstates with different principle quantum number n but identical angular momentum and azimuthal quantum numbers l and m, are easily characterized using SSFI. However, SSFI is not as effective for dissecting the state composition of more complicated wave packets that are superpositions of states with different n, l, and m. Even if the state compositions can be measured, there is still the difficulty of creating an appropriate reference packet whose quantum state is completely characterized. For the cross-correlation method to be successful every eigenstate in the target superposition must also be represented in the reference packet. Therefore, target wave packets with complicated eigenstate distributions require similarly complex reference packets whose quantum state may not be known with high confidence.

In this paper we demonstrate an alternative method for complex amplitude retrieval based on an analysis of time-dependent probability distribution measurements. Like the cross-correlation technique, the probability distribution method is more robust if the eigenstate composition of the wave packet has been ascertained through SSFI or some other method. However, neither phase coherence between pump and probe pulses nor a well characterized reference wave packet are required.

Consider an arbitrary wave packet

$$\Psi(\vec{r},t)=\sum_j A_j\psi_j(\vec{r})e^{-i(E_jt-\phi_j)}, \tag{1}$$

where the A_j are the real, time-independent amplitudes of the constituent eigenstates $\psi_j(\vec{r})$ with energies E_j and constant phase ϕ_j. The sum extends over all quantum numbers and atomic units are used throughout. Note that the wave function could be equally well represented in momentum space by substituting $\vec{p}$ for each instance of $\vec{r}$ in the previous and following equations. The time-dependent probability distribution is

$$P(\vec{r},t)=\sum_{j,k} A_jA_k\psi_j(\vec{r})\psi_k(\vec{r}) \times\exp\{i[(E_k-E_j)t-(\phi_k-\phi_j)]\}, \tag{2}$$

where we have assumed real eigenfunctions ψ_j without loss of generality. The phase difference $\Delta\phi_{kj}=(\phi_k-\phi_j)$ between any two constituent eigenstates can be recovered from the Fourier transform, $S(\vec{r},E)$, of the measured probability distribution $P(\vec{r},t)$. $S(\vec{r},E)$ consists of a series of spikes at the difference frequencies $E=\Delta E_{kj}=E_k-E_j$. The phase of $S(\vec{r},\Delta E_{kj})$ evaluated at the center of a particular resonance feature is related to the phase difference between states ψ_k and ψ_j, $\Phi_{kj}=\mp\Delta\phi_{kj}=\mp(\phi_k-\phi_j)$. The choice of + or − as the multiplicative factor depends on the sign of the product $\psi_j(\vec{r})\psi_k(\vec{r})$, a quantity which is accurately known for Rydberg eigenstates. The minus sign applies to positive eigenstate products.

For a superposition of N eigenstates, $N-1$ phase differences are measured. However, since the quantum state of the wave packet can only be determined up to a multiplicative phase factor, the phase ϕ_i of one of the eigenstates, ψ_i, can be arbitrarily set to zero. This selection, combined with the measured phase differences, defines the phases of the other $N-1$ states in the packet. Note that no physical observable is affected by the choice of phase for eigenstate ψ_i.

In principle, the real eigenstate amplitudes, A_j, can also be determined from $S(\vec{r},E)$. The ratio of any two amplitudes, A_j/A_k, is directly proportional to $S(\vec{r},\Delta E_{ji})/S(\vec{r},\Delta E_{ik})$. The constant of proportionality depends only on the eigenstate products $\psi_j\psi_i$ and $\psi_i\psi_k$ which are known. However, in practice, experimental noise in the measured probability distributions have a much greater effect on the peak amplitudes $S(\vec{r},\Delta E)$ than on the phase and energy differences, $\Delta\phi$ and ΔE. This amplitude noise makes an accurate determination of the eigenstate amplitudes very difficult. However, if the real eigenstate amplitudes A_j have been independently measured (e.g., using SSFI), then the phase retrieval described above completes the measurement of the quantum state of the wave packet.

Of course, the distribution function $P(\vec{r},t)$ cannot be measured exactly. Instead, the probability for finding the wave packet within some volume ΔV between $\vec{r}$ and $\vec{r}+\vec{\delta r}$ is determined. Fortunately, integration of $S(\vec{r},E)$ over ΔV affects the amplitudes $S(\vec{r},\Delta E_{kj})$, but does not alter the phases $\Delta\phi_{kj}$. Therefore, the phase measurement is not compromised. It is also worth noting that the probability measurement over any finite volume ΔV contains phase-difference information for every pair of constituent eigenstates. As a result, in many cases only a small fraction of the probability distribution, e.g., the small radius portion near the ionic core, must be measured to accurately determine all of the phase differences.

II. RESULTS

We now demonstrate the complex phase-retrieval method using previously published data from two radial wave packet [4] experiments. In the first experiment, time-resolved isolated core excitation (TRICE) was used to measure the time-dependent radial probability distribution of a coherent superposition of $4snd$ states in calcium [9]. In the second experiment, half-cycle pulse ionization was utilized in the impulsive momentum retrieval (IMR) method to determine the momentum space probability distribution of a similar $4snd$ wave packet [16]. In both cases, the laser pulses used to excite the wave packet were frequency chirped and the constituent eigenstate amplitudes were measured using SSFI. The details of the experiments can be found elsewhere [9,16] and are not reproduced here. The measured constituent eigenstate amplitudes and recovered phases for the wave packets studied in the TRICE and IMR experiments are shown in Figs. 1 and 2, respectively. Clear phase variations across the eigenstate distributions are evident in both wave packets.

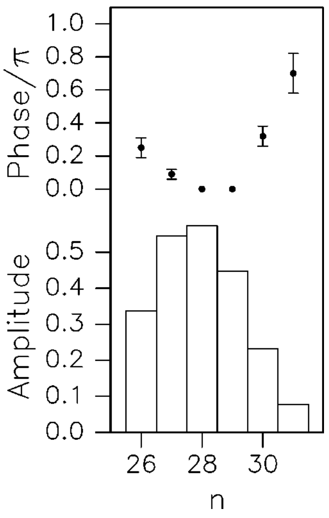

FIG. 1. Measured amplitudes and phases of the constituent eigenstates in a $4snd$ radial wave packet in calcium. The real amplitudes were measured using SSFI and the phases were obtained from Fourier transforming radial probability distribution measurements from previous TRICE experiments. Since only phase differences are measured, the quantum state can only be known up to a multiplicative phase factor. Therefore, we define the phase of the $4s28d$ eigenstate to be zero.

In the TRICE experiment, phase retrieval was performed using portions of the radial probability distribution corresponding to the inner and outer turning points of the wave-packet motion (see Fig. 3). Since the $4snd$ eigenfunctions all have the same radial phase at small r, each eigenstate product contributes a + sign to the phase measurement at small radius. Conversely, at large r, the radial eigenfunctions are 180 ° out of phase and the eigenstate product is negative. As a consequence, the phases Φ_{kj} retrieved from the two data sets have a relative π phase shift. This phase shift has been accounted for in the plot of $\Delta\phi_{kj}$ shown in Fig. 1. The error bars in Fig. 1 indicate the differences in the phases obtained using the small and large radius data, respectively. Errors in the phase determination are only significant for states with small amplitude whose frequencies do not figure prominently in the Fourier transform data (see Fig. 4).

The IMR measurements from the second experiment provide momentum distributions in Cartesian rather than spherical coordinates. As a result, care must be taken to accurately assess the appropriate sign of the eigenstate products that can

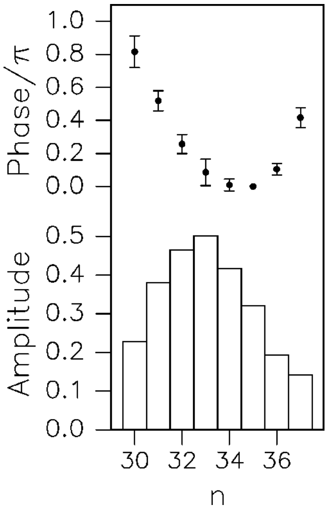

FIG. 2. Measured amplitudes and phases of the constituent eigenstates in a $4snd$ radial wave packet in calcium. The real amplitudes were measured using SSFI and the phases were obtained from Fourier transforming momentum-space probability distribution measurements from previous IMR experiments. Since only phase differences are measured, the quantum state can only be known up to a multiplicative phase factor. Therefore, we define the phase of the $4s35d$ eigenstate to be zero.

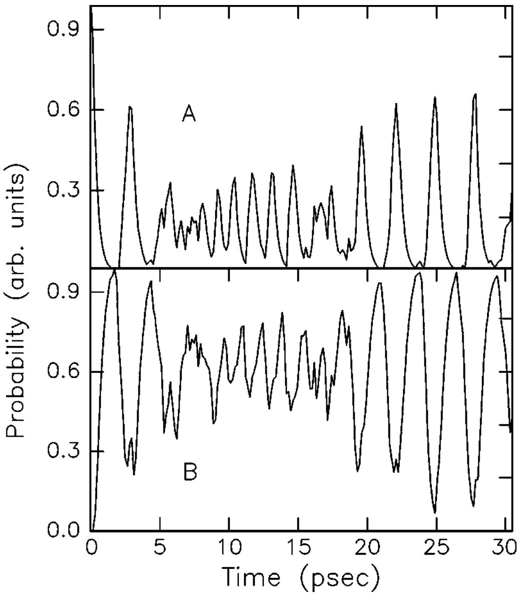

FIG. 3. Measured probability for finding the wave packet described in Fig. 1 (a) at small radius near the ion core and (b) at large radius near the classical outer turning point.

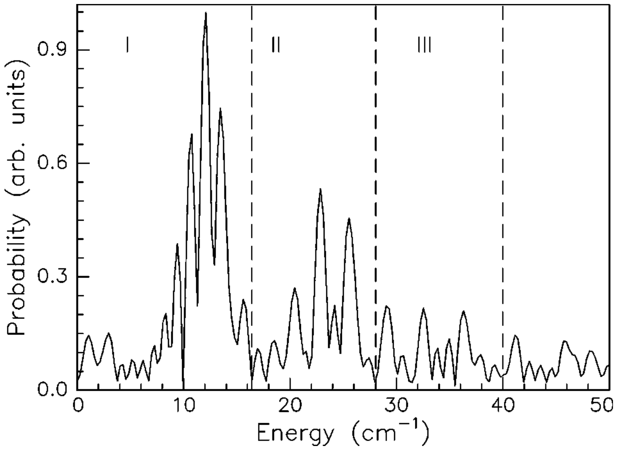

FIG. 4. Discrete Fourier transform of the small radius probability data shown in Fig. 3(a). The vertical dashed lines separate regions I, II, III, which indicate energy splittings between $4snd$ levels with $\Delta n=1,\ 2$, and 3, respectively. The phases shown in Figs. 1 and 2 are associated with region I peaks.

contribute a π phase shift to the measured phases $\Delta\phi_{kj}$. For example, the probability distribution evaluated near zero momentum along the z axis, $p_z=0$, depends on electron probability with very small total momentum (i.e., near the outer turning point) as well as probability with large momentum along a different axis (i.e., moving with high velocity near the nucleus along $\hat{x}$ or $\hat{y}$). Unfortunately, the eigenfunction products $\psi_k(\vec{p})\psi_j(\vec{p})$ from different volumes in momentum space have different signs. As a result, it can be difficult to determine the correct sign of $\Delta\phi_{kj}$ from the measurement of Φ_{kj}.

To make the phase evaluation as straightforward as possible, we only consider the high momentum part of each distribution. Large momentum components along any Cartesian axis imply large radial momentum as well. Since the phases of the relevant momentum-space eigenfunctions are identical at large radial momentum, each eigenfunction product, $\psi_k(\vec{p})\psi_j(\vec{p})$, in Eq. (2) contributes a + sign to the measured phase $\Phi_{kj}=-\Delta\phi_{kj}$. Figure 2 shows the average phase retrieved from an ensemble of phase determinations. Typically, twelve phase measurements were obtained for each eigenstate. Each determination was made using a different high momentum component of the experimental distributions along either the $\hat{x}$ and $\hat{z}$ directions. The error bars in Fig. 2 represent standard error of the mean for the ensemble.

Once determined, the constituent eigenstate phases and amplitudes can be used in conjunction with detailed knowledge of the eigenfunctions and energies to reconstruct the actual wave-packet evolution. A wave-packet movie, composed of snapshots of the wave packet's evolving Wigner distribution, is particularly insightful. A less attractive alternative which will suffice for the paper journal medium is a density plot of the time-dependent coordinate and momentum space probability distributions. The measured wave-packet distributions obtained from the TRICE and IMR data are shown in Figs. 5 and 6, respectively. Precise knowledge of the eigenfunctions and eigenenergies makes it possible to reliably track the wave-packet motion from the measurement of only a few complex amplitude parameters.

III. DISCUSSION

As noted in the Introduction, phase recovery from probability distributions has an advantage over the cross-correlation technique [14] in that complicated reference wave packets are not required for accurate phase retrieval.

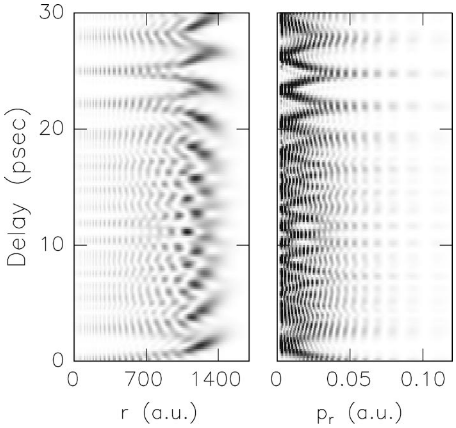

FIG. 5. Density plots of the measured time-dependent probability distribution for the 4*snd* radial wave packet described by Fig. 1 in coordinate and momentum space. Dark denotes a region of high probability. The time interval displayed matches the delay range over which the original TRICE measurements were made.

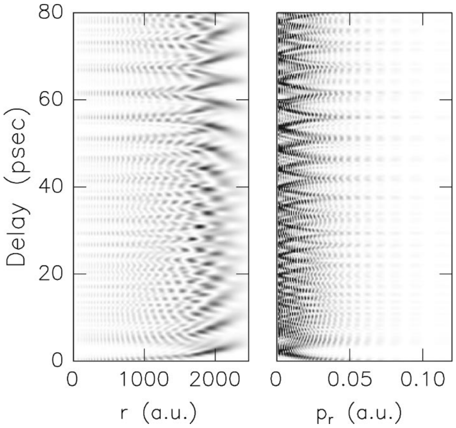

FIG. 6. Density plots of the measured time-dependent probability distribution for the 4*snd* radial wave packet described by Fig. 2 in coordinate and momentum space. Dark denotes a region of high probability. The time interval displayed matches the delay range over which the original IMR measurements were made.

Unfortunately, the probability method has a different technical limitation. Specifically, the technique relies on identifying energy differences to define specific phase differences between eigenstates. If different pairs of states have equal or nearly equal energy splittings, this determination becomes difficult in practice. In principle, by measuring the phase differences using data from different spatial volumes, one could sort out the phase differences corresponding to each eigenstate pair. However, this approach is unlikely to yield accurate results for a large number of degenerate energy splittings.

The real advantage of the probability-based phase-recovery method lies in the characterization of wave packets that are linear superpositions of several principal and low angular momentum quantum numbers in nonhydrogenic atoms. The non-negligible quantum defects of low angular momentum states should enable a clear distinction between all relevant energy splittings in the Fourier transform of a measured probability distribution. As a result, extraction of each phase difference should be straightforward. We suspect that such a wave packet would not be easily characterized using the cross-correlation method due to the difficulty in producing a well-known reference packet with multiple angular momentum components.

IV. SUMMARY

In summary, the quantum state of electronic wave packets can be determined from measurements of the wave packet's time-dependent probability distribution and eigenstate composition. The method has been demonstrated for chirped radial wave packets using previously measured coordinate and momentum space distributions. It is worth noting that the momentum distribution data were collected using delay imaging techniques [17]. Delay imaging can be utilized with TRICE, IMR, or short-pulse photoionization to provide the time-dependent probability distribution data required for phase retrieval in a single laser shot. Since SSFI is also a single-shot measurement, real-time evaluation of the quantum state of electronic wave packets should be possible using current technology.

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