Reconstruction of engineered atomic wave functions via phase-dependent population measurements

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A wave-packet interference technique is proposed to measure exotic atomic Rydberg wave functions. For example, the subject wave packet is excited by an arbitrary pulse or pulse sequence. The resulting wave function is probed by a well-characterized pulse that shares a phase relationship with the excitation pulse(s). The observable is the population in each eigenstate after the probe pulse. By making a series of such measurements, varying the phase between excitation and probe pulses, the wave function created by the excitation pulse can be inferred. The method is experimentally feasible for atomic systems, and the extension of the method to molecular systems also appears possible. [S1050-2947(97)05309-2]

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A quantum system is fully defined by its wave function or density matrix. However, neither of these is an observable. It is the conflict between the desire to know everything about a system and this inability to make the measurement; that is, the source of the great interest in methods for experimentally extracting the wave function or density matrix of a system. These methods all rely upon preparing n identical systems, and making a sequence of measurements upon these systems. The wave function or density matrix is then inferred from this sequence of experimental observations, e.g., from marginal probability distributions.

The recent theoretical interest in the reconstruction of quantum states was motivated by the seminal work of Vogel and Risken [1]. The original problem was actually posed by Pauli many years ago [2]. Since that time, theoretical investigations have been performed upon spin systems [3], harmonic systems (e.g., quantized fields) [4–6], and bound [7] and free particle systems [8]. Experimental interest in the coherent control or engineering of quantum systems has also spurred interest in this measurement problem. It is now possible to generate engineered wave functions in a multitude of systems [9–14], so that techniques for measuring such exotic states are particularly timely. For example, a Schrödinger cat state has been prepared experimentally in a Rydberg atomic system [14] .

In Ref. [1] the Wigner function (which bears the same information as the density matrix) is linked to a set of measured probability distributions, $P_\phi(X_\phi)$, of quadrature amplitudes by an inverse Radon transform. Sufficient information to reconstruct the Wigner function is obtained by sampling the phase from 0 to π. This type of reconstruction of the Wigner quasiprobability distribution from such marginal probability distributions is commonly known as quantum tomography. Currently this type of tomography has been employed experimentally on such systems as quantized radiation fields [15], trapped ions [16], vibrational modes of molecules [17], and free particles [18]. Theoretical studies of anharmonic systems have just recently been performed [7,19,20]. At present, the experimental application of tomographic techniques to anharmonic systems has not been fully accomplished.

Of particular interest to this paper is the recent theoretical effort linking wave functions of atomic wave packets to their motion. Leonhardt and Raymer [19] studied the reconstruction of wave packets in arbitrary one-dimensional systems. Their observable is the positional probability distribution $P(x,t)$. From the Fourier transform of $P(x,t)$, the elements of the density matrix may be extracted with the appropriate sampling functions. This technique does pose some experimental challenges, e.g., the temporal and spatial sampling of $P(x,t)$ are not trivial problems. Experimental progress has been made in measuring $P(p,t)$, the momentum probability distribution, for a Rydberg atom wave packet [21]. Although it has not been demonstrated, it is likely that a similar reconstruction of the wave function could be accomplished from such momentum data.

The reconstruction of exotic atomic Rydberg wave packet states, such as the Schrödinger cat state, is the primary focus of this paper. We propose an experimentally feasible scheme combining Ramsey interference [22] between wave packets with state-selective field ionization (SSFI) [23] to realize the reconstruction of the wave-packet state. These are well-established experimental techniques, but their combination has surprising power. The reconstruction technique requires only a small number of measurements and these measurements may be made with a high degree of quantum efficiency. The extension of this reconstruction technique to molecular systems also appears possible. This scheme utilizes a recently developed formalism to analyze the interference of wave packets [24].

Wave packets in the hydrogenic Rydberg atomic system have been studied experimentally over the past decade. A few examples are Refs. [21,25–32]. Typically, a coherent superposition of Rydberg states is excited by a short optical pulse, creating a wave packet that can display both classical and quantum-mechanical features. The creation of more exotic wave packets can be accomplished by a programmable pulse [9], autoionization [33], or through the Ramsey interference of multiple pulses [14,24,34]. This last method will serve, in this paper, as the basis for a concrete example of our reconstruction technique. The essence of this Ramsey interference is that the ultimate populations of the Rydberg states are sensitively dependent upon the optical phase and upon the motion of the wave packets. Thus, by introducing a

pulse with a specific optical phase, at a suitable point in the evolution of the already existing wave packet, the amplitudes of the Rydberg states can be controlled in a nearly arbitrary way. This idea has been described with a simple mathematical formalism in our previous paper [24]. This formalism leads directly to the interference technique described here.

The subject wave function is created by a sequence of pulses. For now, we will assume that all the pulses are identical and have a Gaussian shape, $g(t)=(1/\sqrt{2\pi}\sigma)\exp(-t^2/2\sigma^2)$, where σ characterizes the width of the pulse. Generalization of this to an arbitrary excitation pulse shape will be discussed later in this paper. Following this sequence of Gaussian pump pulses, a probe pulse excites a wave packet that interferes with the previously generated wave packets. Here the probe pulse is also taken to be Gaussian in shape, and has a definite phase relationship with the initial pump pulse. This condition on the probe pulse shape may be relaxed somewhat, and will be discussed later in this paper. Following each pump-probe interaction, the population of each Rydberg state is measured by some high-resolution technique (e.g., state-selective field ionization). A few such measurements of the Rydberg state population distribution, in which the relative optical phase between the pump and probe pulses is varied, is sufficient to reconstruct the wave function.

The atomic system may be described by the radial wave function,

$$\Psi(r,t)=a_g(t)\exp(-i\omega_g t)u_g(r)+\sum_n a_n(t)\exp(-i\omega_n t)u_n(t),$$

where a_n and ω_n are the Schrödinger amplitudes and eigenenergies for the Rydberg states and a_g and ω_g are these quantities for the ground state. For convenience, we will set $\omega_g=0$. Under the rotating-wave approximation, the interaction of a pulse (or pulses) with the Rydberg states is given by the Schrödinger equation,

$$\dot{a}_n(t)=-\frac{i}{2}\Omega_n a_g e^{i\Delta_n t}f(t), \tag{1}$$

where Ω_n is the Rabi frequency for the excitation between the ground state and a given Rydberg state n. For Rydberg states, this Rabi frequency is proportional to $n^{-3/2}$. The detuning is defined as $\Delta_n=\omega_n-\omega$, and $f(t)$ is the multipulse envelope function,

$$f(t)=g(t)+\sum_i g(t-\tau_i)\exp(i\omega\tau_i), \tag{2}$$

where the τ_i are the time delays for the individual pulses. We assume the intensity of the laser pulses are sufficiently low so that they may be assumed to be perturbative, in which case the ground-state amplitude remains almost unchanged after the short-pulse excitations. This perturbative condition on the excitation pulse is essential to the reconstruction method described in this paper. If the ground state is altered by a strong interaction effect (e.g., the ac Stark effect), the reconstruction cannot be accomplished by this technique. Fortunately, the common situation in which a weak pulse excites the Rydberg wave packet by a one-photon process from the ground state fulfills this condition. The solution of the differential equations, in this situation, is quite simple. The excited-state amplitudes are given by the Fourier transform of the pulse shape. After the last excitation pulse, a_n is

$$a_n=-\frac{i}{2}\Omega_n\exp\left(-\frac{\Delta_n^2\sigma^2}{2}\right)\left[1+\sum_i \exp[i(\omega+\Delta_n)\tau_i]\right]. \tag{3}$$

By manipulating the phase $\omega\tau_i$ and suitably choosing the delay time τ_i, engineered wave functions such as Schrödinger Cat states can be generated [14,24]. Next we will show how such an engineered state can be reconstructed by a series of measurements.

A simple, but informative reconstruction can be found for a wave packet excited by a single Gaussian pulse. The probability amplitude a_n after that pulse is

$$a_n=-\frac{i}{2}\Omega_n\exp\left(-\frac{\Delta_n^2\sigma^2}{2}\right). \tag{4}$$

Note that a_n is a purely imaginary number. The experiment to reconstruct this wave packet is trivial. All that is required is that we perform SSFI upon the atom, and so retrieve the populations in the various eigenstates. This assumes that the Fourier transform of the probe pulse is real and positive (as is the case for a Gaussian pulse). Also note that the sign for Ω_n is determined by the phase convention for the eigenstates. Therefore the populations $|a_n|^2$ uniquely determine these amplitudes except for a phase factor common to all the Rydberg states.

In the above situation, where the excitation pulse is well characterized, the reconstruction of the quantum state is straightforward. However, when multiple pulses excite a Rydberg atom, the probability amplitude after the last pulse excitation is, in general, a complex quantity. The probability amplitudes a_n cannot simply be determined via a single measurement of either the fringe amplitude resulting from Ramsey interference or of the population as found by SSFI. However, several measurements made upon identically prepared wave packets can regain the amplitudes of the states of the prepared wave packet. Following each excitation sequence, a probe pulse is applied whose phase with respect to the initial excitation pulse is varied. In each measurement, the post-probe population of the various Rydberg states is found by SSFI or some other high-resolution method.

The result of a multiple pulse excitation sequence is given by

$$a_n=-\frac{i}{2}\Omega_n e^{-(\Delta_n^2\sigma^2/2)}(x_n+iy_n), \tag{5}$$

where x_n and y_n are the real and imaginary parts of the bracketed quantity in Eq. (3). Now a probe pulse (identical to the previous pulses) interacts with the atom, producing the new probability $a_n'(\tau)$,

$$a_n'(\tau)=-\frac{i}{2}\Omega_n e^{-(\Delta_n^2\sigma^2/2)}(x_n+iy_n+e^{i\omega\tau}e^{i\Delta_n\tau}), \tag{6}$$

where τ is the delay time for the probe pulse. Again, $a_n'(\tau)$ is not an observable, but the population, $|a_n'(\tau)|^2$, may be measured using SSFI. This population can be expressed as

$$|a_n'(\tau)|^2=\left(\frac{\Omega_n}{2}e^{-\Delta_n^2\sigma^2/2}\right)^2\{(x_n^2+y_n^2+1)+2[x_n\cos(\Delta_n\tau)+y_n\sin(\Delta_n\tau)]\cos(\omega\tau)+2[-x_n\sin(\Delta_n\tau)+y_n\cos(\Delta_n\tau)]\sin(\omega\tau)\}. \tag{7}$$

The terms dependent upon τ, in Eq. (7), each contain terms that oscillate at markedly different frequencies under typical conditions for wave-packet excitation. Typically the oscillation frequency of the wave packet is 1000 times smaller than the optical frequency of pulse. There is one notable exception: Wave packets generated by half-cycle pulses [21] do not fulfill this condition, and this reconstruction technique is not appropriate for analysis of these wave packets. If we consider only the more typical optical excitation, a change of the optical phase, $\omega\tau$, by 2π leaves $\Delta_n\tau$ essentially unchanged. Thus the problem of reconstructing the wave packet is reduced to a series of measurements of the populations, $|a_n'(\tau)|^2$, produced by probe pulses with different but known optical phases. The particular optical phases are chosen that simplify the extraction of x_n and y_n from the data. For example, here we choose a solution that requires measurements at delay times, τ_ϕ, that yield the four specific phases, i.e., $\phi=\omega\tau_\phi=0$, π, $\pi/2$, and $3\pi/2$ [all modulo (2π)]. Thus x_n and y_n are

$$x_n=\frac{|a_n'(\tau_0)|^2-|a_n'(\tau_\pi)|^2}{\Omega_n^2e^{-\Delta_n^2\sigma^2}}\cos(\Delta_n\tau)-\frac{|a_n'(\tau_{\pi/2})|^2-|a_n'(\tau_{3\pi/2})|^2}{\Omega_n^2e^{-\Delta_n^2\sigma^2}}\sin(\Delta_n\tau), \tag{8}$$

$$y_n=\frac{|a_n'(\tau_0)|^2-|a_n'(\tau_\pi)|^2}{\Omega_n^2e^{-\Delta_n^2\sigma^2}}\sin(\Delta_n\tau)+\frac{|a_n'(\tau_{\pi/2})|^2-|a_n'(\tau_{3\pi/2})|^2}{\Omega_n^2e^{-\Delta_n^2\sigma^2}}\cos(\Delta_n\tau). \tag{9}$$

Since $\Delta_n\tau_\phi$ is essentially unchanged for the range of τ's (0 to 2π) the subscript has been dropped when these τ's are associated with Δ_n. As we see, only four measurements are sufficient to retrieve a_n for all the Rydberg states and, in turn, reconstruct the wave function. Note that the specific position of the wave packet in its orbit is not crucial to this method. It is commonly stated that the interaction of a wave packet with a probe pulse is minimal if the probability of finding the wave packet near the nucleus is small during the application of the probe pulse. It is true that the visibility of Ramsey fringes does go to a minimum at this point. However, the amplitudes of the individual eigenstates can be significantly altered even when the wave packet is away from the nucleus. Therefore, the measurement can take place at any position upon the wave packet's trajectory.

In the above example, we assumed that the pump and probe pulses are identical pulses. However this is not a necessary condition for our formalism to work. A generalization to a more arbitrary excitation pulse can be made. There has recently been much interest in tailoring optical pulses so that both the magnitude and phase of the complex scalar field $\Psi(r,t)$ are controlled in a programmable way [9–11]. To generalize our scheme we need only assume that the pump pulse(s) is weak (i.e., the interaction is perturbative) and that it is phase coherent with the probe pulse. The probe pulse is still assumed to be weak and Gaussian in shape. The probability amplitude after the pump pulse is $a_n=-(i/2)\Omega_n(X_n+iY_n)$, where X_n and Y_n, respectively, are the real and imaginary parts of the Fourier-transformed envelope function. The interaction of the probe pulse produces the probability amplitude

$$a_n'=-\frac{i}{2}\Omega_n(X_n+iY_n+e^{-(\Delta_n^2\sigma^2/2)}e^{i\omega\tau}e^{i\Delta_n\tau}). \tag{10}$$

Thus the probability $|a_n'|^2$ is

$$|a_n'|^2=\left(\frac{\Omega_n}{2}\right)^2\{X_n^2+Y_n^2+e^{-(\Delta_n^2\sigma^2)}+2\,e^{-(\Delta_n^2\sigma^2/2)}\times[X_n\cos(\Delta_n\tau)+Y_n\sin(\Delta_n\tau)]\cos(\omega\tau)+2\,e^{-(\Delta_n^2\sigma^2/2)}\times[-X_n\sin(\Delta_n\tau)+Y_n\cos(\Delta_n\tau)]\sin(\omega\tau). \tag{11}$$

The desired values of X_n and Y_n can be obtained in a manner similar to the previous multipulse example, via measurements of the population with appropriately phased probe pulses. More precisely, the values of X_n and Y_n are given by

$$X_n=\frac{|a_n'(\tau_0)|^2-|a_n'(\tau_\pi)|^2}{\Omega_n^2e^{-(\Delta_n^2\sigma^2/2)}}\cos(\Delta_n\tau)-\frac{|a_n'(\tau_{\pi/2})|^2-|a_n'(\tau_{3\pi/2})|^2}{\Omega_n^2e^{-(\Delta_n^2\sigma^2/2)}}\sin(\Delta_n\tau), \tag{12}$$

$$Y_n=\frac{|a_n'(\tau_0)|^2-|a_n'(\tau_\pi)|^2}{\Omega_n^2e^{-(\Delta_n^2\sigma^2/2)}}\sin(\Delta_n\tau)+\frac{|a_n'(\tau_{\pi/2})|^2-|a_n'(\tau_{3\pi/2})|^2}{\Omega_n^2e^{-(\Delta_n^2\sigma^2/2)}}\cos(\Delta_n\tau). \tag{13}$$

To examine the feasibility of the proposed method, we consider the accuracy of the reconstruction of a quantum state of an atomic wave packet produced by the arbitrarily shaped weak pulse described above. One of the primary concerns is the effect of a less than perfect probe pulse. We investigate this problem by examining the reconstruction that is obtained for a probe pulse with a small linear chirp and compare it to that obtained for a transform-limited Gaussian pulse. This chirped pulse is defined $E(t)=(1/\sqrt{2\pi}\sigma)\exp[-(1+ib)t^2/(2\sigma^2)]$. For a chirped pulse with small values of b (keeping only terms linear in b), X_n and Y_n become

$$X_n = X_n^{(0)} - b(\Delta_n^2\sigma^2 - 1)e^{-(\Delta_n^2\sigma^2/2)}Y_n^{(0)}, \quad (14)$$

$$Y_n = Y_n^{(0)} + b(\Delta_n^2\sigma^2 - 1)e^{-(\Delta_n^2\sigma^2/2)}X_n^{(0)}, \quad (15)$$

where $X_n^{(0)}$ and $Y_n^{(0)}$ are the results for a transform-limited Gaussian as defined by Eqs. (12) and (13). The magnitude of the term $(\Delta_n^2\sigma^2-1)e^{-(\Delta_n^2\sigma^2/2)}$ is always less than 1. Therefore, the degree of entanglement of the desired quantities $X_n^{(0)}$ and $Y_n^{(0)}$ is b. That is, if we unknowingly have a pulse with a value of $b=0.01$, the value of X_n obtained for the chirped pulse unknowingly has at maximum 1% of $Y_n^{(0)}$ added to the desired $X_n^{(0)}$. It should be noted that it is experimentally straightforward to create such nearly transform limited pulses [35]. Therefore, it is possible to reconstruct the wave function to a high degree of accuracy. In addition, the ability to characterize ultrafast pulses has recently seen significant advances [36–38]. In fact, transform-limited pulses are not essential to this reconstruction scheme; any perturbative pulse that has been characterized to a similar level can be used as a probe pulse.

A further requirement on the probe pulse is that its bandwidth must equal or exceed that of the pump pulse. The only conditions placed upon the pump pulse are that the interaction is perturbative, and the pulse is coherent with the subsequent probe pulse. An additional experimental requirement is that the measurement of the population must resolve the eigenstates of the system. The technique of SSFI easily satisfies the criteria for resolving the population in weakly bound, low angular momentum Rydberg states. The high quantum efficiency of SSFI is an added advantage. All of these experimental conditions are straightforward to realize in Rydberg-atom wave packet experiments.

To what other systems may this reconstruction scheme be applied? This scheme is aimed at the reconstruction of wave functions of pure states. Systems which lose coherence rapidly (those dominated by spontaneous processes, dephasing, etc.) are not well suited to this interference technique. If the dephasing time is of a similar magnitude to the measurement time, then this method cannot be employed. For a typical wave packet, the measurement time can be a small fraction of the period of the wave packet's oscillation. The system we have been discussing, atomic Rydberg states, have exceptionally slow dephasing processes (roughly 10^6 times longer than the orbital period). Molecular systems typically have shorter dephasing times but they are long enough that many periods of the wave packet's motion can and have been observed [10,11,17,39]. Therefore, there is a significant period of time in which the molecular wave packet is essentially a pure state. This reconstruction scheme also requires that a means for resolving the populations in the various states of the superposition exist. Recent experiments suggest that such resolution is possible for molecular vibrational wave packets [39]. The situation with solid-state systems is less certain. Here the dephasing times are quite short. For example, in the bulk semiconductor the dephasing time for a free exciton is approximately 100 fs [40]. However, in a confining potential, e.g., a one-dimensional quantum well, this dephasing time can be increased to approximately 3 ps [41]. In that work, a pump and a delayed probe pulses were used to produce phase-sensitive control of THz radiation from a polarized electron-hole pair. This retention of coherence suggests that there may be a short time in which the reconstruction could be accomplished for the quantum well system.

The interference technique described in this paper is ideally suited to the study of Rydberg atomic wave packets. We have demonstrated that the reconstruction of the wave function of an atomic Rydberg wave packet requires only a small number of measurements. These phase-dependent population measurements offer a means of determining the wave functions of the programmed or engineered Rydberg atomic wave packets that have recently been generated.

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