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Wave-packet reconstruction in a two-electron atom via impulsive isolated core excitation

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In a two-electron atom, autoionization from an impulsive isolated core excitation is a sensitive probe of a Rydberg electronic wave packet. This probe via the electron-electron correlation allows a quantum measurement of the wave function to be made. In addition to this capability, the measurement process has the advantage that it is independent of the excitation process. An analytical formula describes this probe. This formula is used in conjunction with a nonlinear least-squares algorithm to reconstruct the wave function of this wave packet. The algorithm is relatively insensitive to noise. With 20% added Gaussian noise, the algorithm can still reproduce the amplitudes and phases of the wave function. [S1050-2947(99)50112-1]

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Research on quantum measurement or the reconstruction of the wave function of quantum states has been spurred by growing analytical needs and by the somewhat surprising level of theoretical and experimental success in performing such reconstructions. The accurate measurement of both amplitude and phase of a wave function is of fundamental interest and plays a key role in the area of quantum-state engineering (coherent control) [1]. The seminal work by Vogel and Risken [2] reopened the modern discussion of the reconstruction problem. Since that work, reconstruction techniques have been developed for a variety of systems, including the optical field [3], atomic systems [4,5], and other anharmonic systems [6–11]. Experimental implementations of these techniques have been successfully accomplished for the optical field [12], Rydberg atoms [13], and diatomic molecules [14]. The measurement scheme in this paper uses Rydberg wave packets in atomic systems as its subject. Such wave packets may serve as key elements in the applications mentioned above and also offer test beds for these ideas in more complex systems. In this case, the quantum measurement is being extended to the more complex system of a two-electron atom.

Rydberg wave packets are coherent superpositions of highly excited atomic eigenstates. When these eigenstates differ only in principal quantum number (n), these wave packets are known as radial wave packets. These are routinely created by short pulse excitation of the valence electron in a single-electron atom. A typical wave packet consists of a few states with different n distributed about a large average value $\bar{n}$. These wave packets display classical oscillations with a period of $\tau_{cl}=2\pi\bar{n}^3$ and nonclassical collapses and revivals [15–17]. Such radial wave packets will be used to demonstrate the reconstruction scheme in this paper.

Only recently has a reconstruction technique been proposed [4] and implemented [13] for single-electron Rydberg atomic wave packets. In that work, the wave packet is reconstructed via a probe laser pulse that is coherent with the excitation pulse. In this paper, we propose an incoherent method for retrieving the wave function of a Rydberg atomic wave packet in a two-electron atom. Incoherent, in this context, means that the probe pulse need not have a phase relationship with pump pulse or whatever mechanism that created the Rydberg wave packet. Instead, it is the electron-electron correlation that provides the access to the amplitudes and phases of the states of the wave packet. A major advantage of such an incoherent scheme is that it separates the measurement process from the excitation process. The scheme is not limited to two-electron systems, it may be considered for any system in which some correlation exists; e.g., the correlation between a Rydberg electron and the nuclear motion of a diatomic molecule. In fact, the two-electron atomic system is chosen as a test case for studying quantum measurement in systems where correlations play a significant role. In addition, we have developed a reconstruction algorithm that incorporates a nonlinear, least-squares method that is relatively insensitive to the presence of noise and to near-degeneracy problems.

There is a large volume of work on wave packets in two-electron atoms (a few examples are [18]). The work performed by Jones [19] is of particular interest to this paper. One of the major results of this paper is to provide a straightforward analytical formalism that can reproduce, in detail, the experimental results of that work and that of an earlier work by Story *et al.* [20]. The data produced by this type of probe [19] are of sufficiently high quality that the reconstruction algorithm that is the main focus of this paper should be feasible experimentally.

In a two-electron atom, the correlation between the two valence electrons opens up a wide range of new physical effects. Notably when two electrons are doubly excited the process of autoionization becomes possible. The effects of autoionization and of an excited core configuration can significantly alter the evolution of the wave packet. For example, electron-electron correlations have been shown experimentally to modify the dynamics of wave packets [21]. It was also shown that the autoionization signal can be used as a sensitive probe of the location of a Rydberg wave packet [19].

Figure 1 provides an energy-level diagram indicating the proposed interaction in this reconstruction scheme. The subject wave packet is excited by some mechanism (e.g., a short laser pulse). It is a superposition of a few highly excited bound states. The time of excitation and the range of states involved in the wave packet are the only detailed knowledge

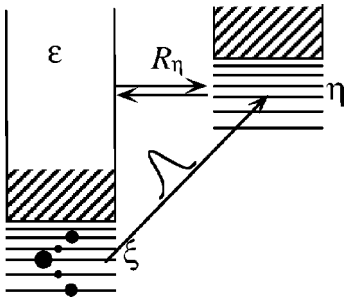

FIG. 1. Energy-level diagram describing the interaction of ICE probe with the bound wave packet. The ICE probe occurs at some delay time τ after the subject wave packet is created, and transfers only a small amount of population to the autoionizing Rydberg series. This population autoionizes through a channel interaction governed by the matrix element R_η. The total autoionization signal, as a function of τ, is the data for the proposed reconstruction scheme.

needed about the excitation process. The Rydberg states are highly excited, so that to first approximation the core is left as if the outer electron were ionized. This situation is sometimes known as isolated core excitation (ICE). The remaining valence electron (or core electron) can be further excited with a laser tuned near a resonance of the nearly ionized core. In this paper, a short pulse drives this resonance of the core electron. This driving couples the bound Rydberg series of levels to the autoionizing levels. However, the probability of reaching the autoionizing series depends upon the delay time τ between the excitation process and when this probe pulse occurs. Semiclassically this can be associated with the overlap of the Rydberg wave packet with the core electron. The inner electron is always near the core, but the wave packet oscillates in the radial direction so that the autoionization process is sensitive to the location of the wave packet. This was demonstrated experimentally in [19]. We will show below that from a series of measurements of the total autoionization signal for several values of τ the wave function of the wave packet may be reconstructed. Note that autoionization is an efficient, resonant process. This allows the probe pulse to be weak, which reduces the possibility that it will drive undesirable processes, e.g., ac Stark shifts.

The formalism described in [22] is used to describe the interactions depicted in Fig. 1. There are two times of significance in this problem. The first is the time at which the bound-state wave packet was excited. The second is the time that the probe laser occurs. We will use an interaction picture to focus on the probe portion of the process. In this picture, the time of the probe pulse is zero. The probability amplitudes for the autoionizing Rydberg series are labeled as $A_\eta(t;\tau)$, and for the bound series as $B_\xi(t;\tau)$ (η and ξ are the quantum numbers for these two Rydberg series). Note that these amplitudes are dependent upon the parameter τ, which is the time between the excitation of the wave packet and when the probe pulse occurs. Free precession relates the bound-state amplitudes at the time the wave packet is created with the bound-state amplitudes at the time of the probe pulse. That is, $B_\xi(t=0;\tau)=|b_\xi|e^{i\phi_\xi}\exp(-iE_\xi\tau)$, where $|b_\xi|$ and ϕ_ξ are, respectively, the magnitudes and phases of the amplitudes of the bound states at the time the subject wave packet was created. With these simplifications, the Schrödinger equations describing this system may be written as,

$$i\dot{A}_\eta(t;\tau)=\frac{1}{2}\sum_\xi S_{\eta\xi}g(t)B_\xi(t;\tau)e^{-i(\Delta+E_\xi-E_\eta)t} - i\pi R_\eta\sum_{\eta'} R_{\eta'}A_{\eta'}(t;\tau)e^{-i(E_{\eta'}-E_\eta)t}, \quad (1)$$

$$i\dot{B}_\xi(t;\tau)=\frac{1}{2}\sum_\eta S_{\eta\xi}g(t)A_\eta(t;\tau)e^{i(\Delta+E_\xi-E_\eta)t}. \quad (2)$$

The energies E_ξ and E_η are $E_j=-1/[2(j-\delta_j)^2]$ ($j=\xi,\eta$). The δ_j are quantum defects. Δ is the detuning of the laser frequency from the isolated core resonance. $S_{\eta\xi}=D_{\eta\xi}(2\sqrt{\eta\xi}\sin[\pi(\eta-\xi)])/[\pi(\eta^2-\xi^2)]$ is the Rabi frequency for the core excitation, determined by the matrix element of the dipole operator and the electric-field amplitude of the light. The channel interaction coupling the upper series and the continuum is governed by $R_\eta=V_0/\eta^{3/2}$, where V_0 specifies the strength of the channel interaction for a particular atomic system. The pulse envelope of the probe pulse is given by the real function $g(t)$.

A complete theoretical description of the ICE autoionizaton process requires a numerical treatment; e.g., a numerical integration of Schrödinger's equations of motion. However, an analytical solution is possible when the probe pulse is sufficiently short and weak. This impulse approximation allows the second term of the right-hand side (rhs) of Eq. (1) to be dropped, provided the probe is short enough that no population is autoionized during the probe. Since the probe laser pulse is also weak, the bound Rydberg-state population is almost unchanged. In this no-depletion limit, when we integrate the rhs of Eq. (1) the quantity $B_\xi(\tau)$ does not depend upon time and so can be pulled out of the integration. Thus, the amplitudes of the autoionizing Rydberg series, right after the probe has occurred, are

$$A_\eta(\tau)=\frac{i}{2}\sum_\xi S_{\eta\xi}B_\xi(\tau)\exp\left(-\frac{(\Delta+\Delta_{\xi\eta})^2\sigma^2}{2}\right). \quad (3)$$

The energy difference $\Delta_{\xi\eta}=E_\xi-E_\eta$ has been introduced. Also, a Gaussian pulse shape has been assumed for the probe pulse [$g(t)\propto e^{-t^2/2\sigma^2}$]. However, this particular shape is not essential to the formalism (any well-characterized short pulse is sufficient).

On a short time scale the autoionization of the population in the autoionizing series to the continuum is complicated [22]. Interference between the various states of the wave packet occurs as they couple to the continuum. However, for times much longer than τ_{cl} the free precession of the various states washes out these interference effects and essentially all the population in the autoionizing series has decayed to the continuum. Therefore, the total number of autoionized electrons that may be detected, e.g., by a microchannel plate (MCP) detector as in [19], is proportional to the total population in the autoionizing states. This electron signal $S(\Delta,\tau)$ is given by summing the population in the autoionizing states that was generated by the probe pulse; so from Eq. (3),

$$S(\Delta,\tau)=\sum_{\eta}|A_{\eta}|^2=\frac{1}{4}\sum_{\xi,\xi'}O_{\xi,\xi'}B_{\xi}(\tau)B^{*}_{\xi'}(\tau), \qquad (4)$$

where

$$O_{\xi\xi'}(\Delta)=\sum_{\eta}S_{\eta\xi}S_{\eta\xi'}e^{-[(\Delta+\Delta_{\xi\eta})^2+(\Delta+\Delta_{\xi'\eta})^2]\sigma^2/2}. \qquad (5)$$

The matrix elements $O_{\xi,\xi'}$ are determined by the system and the pulse parameters. Note that the sum on η must range over a sufficient number of states. This is identical to requiring the bandwidth of the probe pulse to equal or exceed that of the excitation mechanism. This formalism has reproduced all the major features of the experimental results obtained in [19,20].

The amplitudes of interest for the reconstruction of the subject wave packet are the amplitudes of the bound states at the time of excitation. These amplitudes are given by $B_{\xi}(\tau)=|b_{\xi}|e^{i\phi_{\xi}}\exp(-iE_{\xi}\tau)$. They are inserted into Eq. (5), and the resulting form naturally separates into dc and ac terms, $S(\Delta,\tau)=S_{dc}(\Delta)+S_{ac}(\Delta,\tau)$:

$$S_{dc}(\Delta)=\frac{1}{4}\sum_{\xi}O_{\xi\xi}|b_{\xi}|^2, \qquad (6)$$

$$S_{ac}(\Delta,\tau)=\frac{1}{2}\sum_{\xi<\xi'}O_{\xi\xi'}|b_{\xi}|\,|b_{\xi'}|\cos(\phi_{\xi}-\phi_{\xi'}+\Delta_{\xi'\xi}\tau), \qquad (7)$$

where $\Delta_{\xi'\xi}=E_{\xi'}-E_{\xi}$. The reconstruction scheme could make use of either or both the detuning Δ and the time delay τ for retrieving the wave function. However, we find that the detuning may be fixed, and a series of measurements of $S(\tau)$ at several values of τ is sufficient.

Reconstruction requires the retrieval of both the magnitudes and phases of the bound-state amplitudes. The measurement of the probability or the magnitude of each state is a solved problem. State-selective field ionization (SSFI) [23] can determine the population of the Rydberg states with a high degree of quantum efficiency. However, the matrix elements $O_{\xi\xi'}$ may only be known up to a global factor due to an imprecise knowledge of the magnitude of the dipole moment matrix elements or of the intensity of the ICE pulse. To deal with this uncertainty, a global factor is included in the analysis, so that $O_{\xi\xi'}=\gamma\Lambda_{\xi\xi'}$. A fit of S_{dc} gives the global factor γ.

The remaining step in the reconstruction is to find the phases. The phases of the subject wave packet are retrieved by fitting $S_{ac}(\tau)$. That is, the phases of each state are the variables of the fit. For an N-state system we will have $N-1$ independent variables, since there exists an arbitrary global phase (the phase of the first state is chosen as 0). The relationship between S_{ac} and the phases is explicitly nonlinear; thus the linear methods used in [24] cannot be applied directly. Instead a nonlinear least-squares method, known as the Levenberg-Marquardt method [25], is used. This is an iterative method. A χ^2 merit function compares a trial solution to the data and iteratively improves the trial solution until the merit function stops decreasing.

TABLE I. Amplitudes and phases of the subject wave packet. The specified amplitudes and phases are indicated by the superscript (sp). The reconstructed amplitudes and phases are indicated by the amplitude of Gaussian noise included to approximate the experimental noise. The Gaussian noise has amplitudes equal to 0.1 or 0.2 times the signal, $S_{ac}(\Delta,\tau)$, and has a standard deviation of 1. A total of 60 evenly spaced samples were used in the reconstruction.

ξ	$\lvert b_{\xi}\rvert^{(sp)}$	$\phi_{\xi}^{(sp)}$	$\lvert b_{\xi}\rvert^{(0.1)}$	$\phi_{\xi}^{(0.1)}$	$\lvert b_{\xi}\rvert^{(0.2)}$	$\phi_{\xi}^{(0.2)}$
28	0.37	0	0.36	0	0.34	0
29	0.23	1.35	0.23	1.34	0.22	1.32
30	0.74	−0.27	0.75	−0.27	0.74	−0.27
31	0.20	1.40	0.19	1.37	0.17	1.33
32	0.48	0.07	0.46	0.07	0.44	0.08

Table I displays the test results. The test takes place upon a wave packet that is a superposition of five eigenstates. The state chosen is a Schrödinger cat state that is a superposition of two Gaussian wave packets separated by one-half of a classical period, i.e., $\psi(\vec{r},t)+\exp[i(\pi/4)]\psi(\vec{r},t-(1/2)\tau_{cl})$. The eigenstates that make up this wave packet have nontrivial distributions of magnitude and phase. A preliminary SSFI experiment measures the magnitudes. However, this information does not need to be known precisely. The reconstruction algorithm converges to the correct values of magnitude and phase, even with a randomly chosen 20% error in the initial guesses for the magnitudes. The initial trial phases are chosen randomly since no *a priori* knowledge exists for the phases of the states.

Only a relatively small fraction of the time evolution of the wave packet needs to be sampled for the reconstruction process. In the test, the sampled data are taken from an S_{ac} constructed from the magnitude and phases of the wave packet. The samples are evenly spaced and must be sufficiently dense to resolve the highest frequency oscillations possible in S_{ac}. This maximum frequency is defined by the width of the superposition of states that forms the wave packet. This example, where the width is roughly five states, requires approximately ten points per classical period of the wave packet ($\tau_{cl}=2\pi\bar{n}^3$). These samples are taken from a delay time window whose width is equal to 1/2 of the revival time, $(\bar{n}/6)\tau_{cl}$. Samples taken outside this range provide information that is largely redundant. For $\bar{n}=30$, this results in roughly 50 samples of S_{ac}. From such a perfect sample, the iterative process retrieves magnitude and phase with a high degree of precision. Of course, it is expected that experimentally sampled data would include some noise. The addition of 10% and 20% Gaussian noise to the signal S_{ac} yields a maximum error of 7% and 15%, respectively, in any of the reconstructed magnitudes and phases. Thus, this reconstruction algorithm appears to tolerate a significant amount of noise, which suggests that it can be realistically used to handle experimental data; e.g., those in [19]. This robustness can be attributed to the sensitive dependence of S_{ac} on the nonlinear function of the phase. This sensitive dependence also speeds the convergence.

Alternative fitting algorithms exist. For example, the terms $|b_{\xi}|\,|b_{\xi'}|\cos(\phi_{\xi}-\phi_{\xi'}+\Delta_{\xi'\xi}\tau)$ can be grouped and treated as the independent term. In that case, the fit may be

accomplished by a linear least-squares method similar to that used in [24] This method has the advantage of requiring only a matrix inversion to accomplish the fit. For the perfectly sampled data, this linear algorithm also yields the correct values for $|b_\xi|$ and ϕ_ξ. However, when small amounts of noise are introduced ($\sim 0.01\%$), this method fails to produce reasonable results. This critical requirement on the precision of the data is due to the nearly singular matrix involved in this least-squares method. The singularity can be traced to the near degeneracy of the energy differences between the neighboring states of the wave packet. The nonlinear least-squares method is not sensitive to this degeneracy problem. Such degeneracy problems appear in a wide variety of systems.

Specialized quantum states are generated in many systems [26], so that techniques for measuring these designed states are timely. The state reconstruction scheme proposed in this paper extends quantum measurement to the more complex system of the two-electron atom. The incoherent nature of the probe allows the designed wave packet to be produced by any mechanism (no weak-field limitation). This significantly broadens the possible applications of this method. It does not require a phase-sensitive measurement to perform the reconstruction, and this should ease experimental implementation. Finally, the tolerance of noise is a further advantage.

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