Scheme for a coherently controlled pulsed electron gun

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When the outer electron of an alkali atom in an electric field is laser excited just above the field-induced ionization threshold, the electron ejection will not be instantaneous. Calculations show there are a number of energy regions where, upon short-pulse laser excitation, the atom will eject a train of electron probability pulses with the pulses being nearly equally spaced in time. This system can be the source of a picosecond pulsed electron gun. We show that it is possible to coherently control the electron pulse frequency *while* the pulse train is being emitted. © 1998 Optical Society of America [S0740-3224(98)03201-9]

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1. INTRODUCTION

Recent work has focused on the study of the time-dependent electron emission of highly excited alkali atoms in a strong electric field using experimental[1–3] and theoretical[4,5] tools. The measurements on these Rydberg atoms were performed by monitoring the time-dependent flux of electrons that are naturally ejected from the atom. The ejection occurs naturally because the electron has enough energy to travel over the classical barrier to escape. However, because the energy is only slightly above this barrier, the electron wave is trapped near the atom for times of the order of picoseconds. Oscillatory structures in the ejected electron flux arise from the interference of the different energy components of the electron wave.

In this paper we show that alkali atoms in an electric field can be excited in such a way that a very long train (>10) of electron pulses will be ejected with the pulses having nearly uniform width and time separation. This system may be interesting from a practical angle: it could possibly be used as a source in a pulsed electron gun (see Fig. 1). Just as a pulsed laser produces a series of light pulses with a high repetition rate, the alkali atom in a static field can produce coherently controlled electron probability pulses at a high repetition rate. For the systems we have examined the pulse width can be 5 ps and the time between pulses can be 15 ps. The period of the pulses can be varied from 15 ps to 75 ps, allowing a large tuneability in frequency. In contrast to short-pulse electron guns based on photoactivated metal cathodes[6–9] or scattering of an electron beam by a short optical pulse,[10] the electron source discussed here will produce a series of pulses rather than a single pulse. Moreover, the bursts of probability within the pulse train are coherent and can be manipulated while the emission is taking place. We present calculations demonstrating that such a coherent train of electron pulses can indeed be produced by photoexcitation of autoionizing Rydberg states.

2. TIME STRUCTURE OF ATOMIC ELECTRON EMISSION

The dynamics of autoionizing Rydberg states may be fruitfully discussed from both a classical and a quantum point of view. The classical hydrogen atom in a static field evolves as follows: (1) An electron moves out from the nucleus with an energy, E. (2) If the angle between the electron's initial velocity vector and the field direction is smaller than a separatrix angle, which is the maximum angle over which the electron can escape classically, the electron will leave the region near the nucleus and travel downfield. (3) If the initial angle is larger than the separatrix angle, the electron will be trapped forever in the region near the nucleus because this system separates in parabolic coordinates. However, the presence of core electrons in alkali atoms breaks the parabolic symmetry near the nucleus. Each time the electron returns to within a few Bohr radii of the nucleus, the orbit will precess, and the electron's trajectory may now be at an angle smaller than the separatrix angle. Thus it can leave the atom. Therefore one might expect pulses of electrons, with the time between the pulses related to the time required for the electron to return to the core region. The highly excited electron exhibits two types of motion: (1) a radial oscillation from the inner turning point to the outer turning point, of the orbit, and (2) an oscillation of the angular momentum from its initial low l value all the way up to $l = n - 1$ and back. Core penetration, resulting in scattering in the direction of the saddle point, is only possible whenever the angular momentum of the electron is small [because the distance of closest approach is $r_{min} \sim l(l + 1)$] and the electron is at the inner turning point of its radial trajectory.
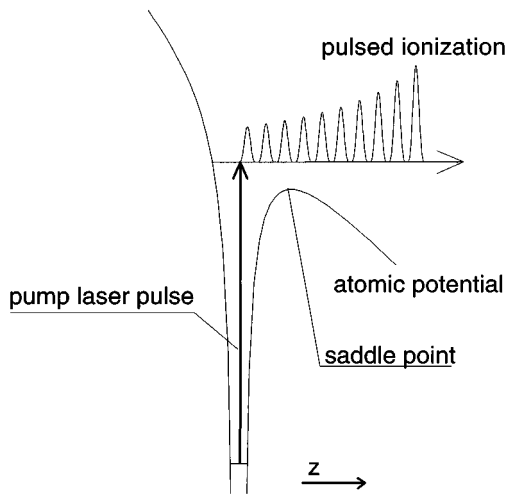
Fig. 1. Schematic representation of a coherent electron gun. A short optical pulse excites a sample of gas-phase atoms in an electric field to an autoionizing wave packet. The electron ejection after the laser excitation occurs in bursts. The time separation of the coherent electron wave packets is determined by the angular-momentum oscillation time.
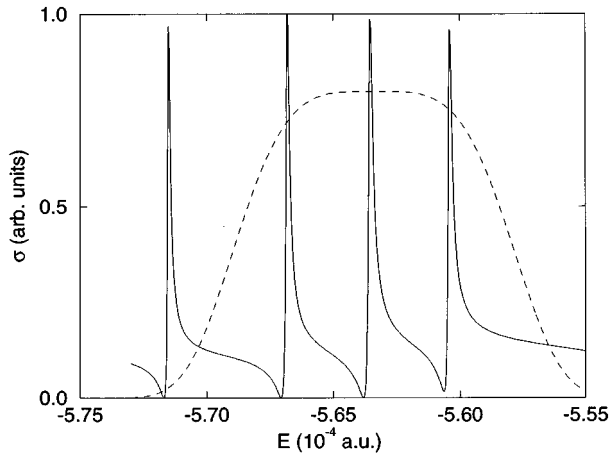

Fig. 2. Solid curve: the photoionization cross section of Na in a static 1.2-kV/cm electric field with the laser polarized perpendicular to the static field direction. Dashed curve: spectrum of the laser pulse.

The quantum description is very similar, yet interference effects can modify the emerging times in a qualitative manner.[5] In the quantum description the wave function for distances larger than a few Bohr radii is a superposition of functions in parabolic coordinates. The coupling between the different channels arises in alkali atoms through the phase shift of the radial function compared with the radial function for a hydrogen atom.[11,12] This coupling allows states to decay more quickly above the classical ionization threshold because it allows electron waves to escape the atom by tunneling *and* by scattering into channels that are classically open. The time evolution of the electron wave is dominated by the spacings and widths of the resonances in the channels that cannot decay classically. A detector downfield measures an initial pulse of electron probability coming from the part of the electron wave initially excited into open channels. Later pulses of electron probability enter the detec-

tor with time spacings that depend on the inverse of the energy spacing of the resonance states. The connection between the classical and quantum description can be made through the use of WKB and closed orbit theory.[5]

3. COMPUTATION OF THE ELECTRON EMISSION SPECTRA

The computation techniques are along the lines of Refs. 4 and 5. The calculated time-dependent flux of electrons may be obtained in a rather straightforward manner from the time-dependent wave function. The wave packet is generated through the action of a weak pulsed laser so the time-dependent part of the wave function is calculated with first-order perturbation theory. The main difficulty is the calculation of the energy-dependent dipole matrix elements. Another difficulty arises because the usual superposition of energy eigenstates becomes an integration over an energy range since the states that are being excited are in the continuum. However, the superposition of the infinite number of continuum states can be performed with ordinary numerical integration techniques. The experimental detection of the time-dependent flux can be accomplished through the development and use of an atomic streak camera.[13]

In Fig. 2 we show the computed total photoionization cross section as a function of energy for Na in a 1.2-kV/cm static electric field. The Stark states may be classified by the three quantum numbers n_1 (number of nodes in the upfield parabolic coordinate), n_2 (number of nodes in the downfield parabolic coordinate), and m (the angular momentum in the direction of the electric field); the principle quantum number is $n = n_1 + n_2 + |m| + 1$. These states may be classified as 22,2,1 at $E = -5.715 \times 10^{-4}$ a.u., 18,8,1 at $E = -5.668 \times 10^{-4}$ a.u., 20,5,1 at $E = -5.635 \times 10^{-4}$ a.u., and 17,10,1 at $E = -5.604 \times 10^{-4}$ a.u. Note that all of these states come from different n manifolds, which arises because this is the very strong n-mixing regime.

The photon energy is measured relative to the zero field threshold in Na. For this case the laser is polarized perpendicular to the static electric field. The fact that these energy levels are nearly equally spaced means that when they are excited by a $\sim$15-ps laser pulse (that has a bandwidth comparable to the spacing of these levels), a wave packet will be created that has very little dispersion. The difference in the energy spacings is $\sim$3% of the energy spacing. A wave packet constructed from these states will not be bound to the atom forever but will be ejected from the atom because these are autoionizing states.

In Fig. 3 the flux of electrons ejected from the atom is plotted as a function of time. It is clear from this figure that the flux comes in pulses. The pulse period is given by $\tau = 2\pi/\Delta E$, where ΔE is the spacing of the resonances shown in Fig. 2. The time between pulses for this case is $\tau \approx 48$ ps. The width of each electron pulse is roughly τ/N, where N is the number of states excited by the laser; to make sharp electron pulses it is necessary to increase N by finding several states with equal spacing. The number of useful pulses that will emerge from this system depends on how accurately the autoionizing resonances
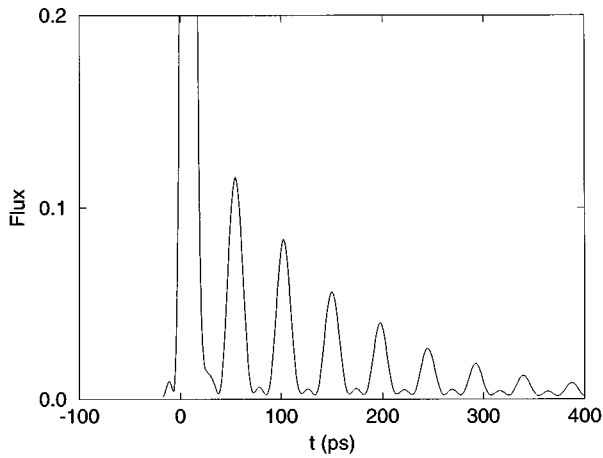
Fig. 3. Electron flux as a function of time for excitation with the laser spectrum shown in Fig. 2.
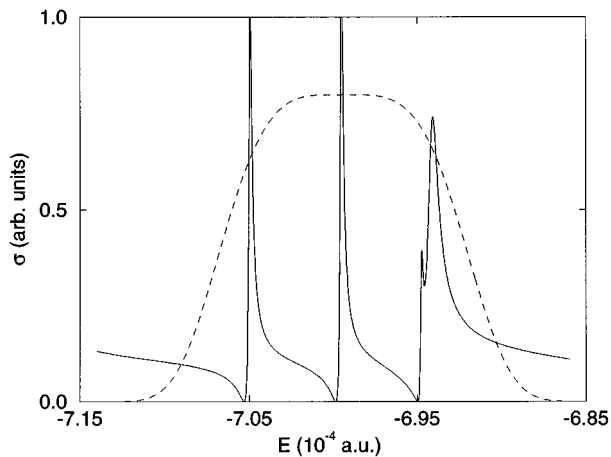

Fig. 4. Solid curve: the photoionization cross section of Na in a static 1.6-kV/cm electric field with the laser polarized perpendicular to the static field direction. Dashed curve: spectrum of the laser pulse.
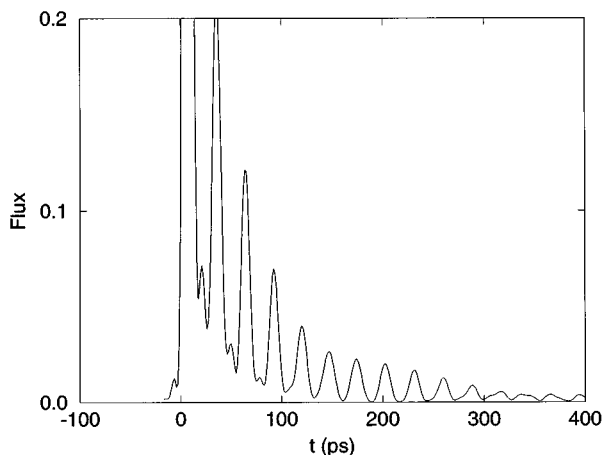

Fig. 5. Electron flux as a function of time for excitation with the laser spectrum shown in Fig. 4.

are spaced and on the lifetimes of the autoionizing states. If the resonances are not equally spaced, dispersion will cause the wave packet to spread as it travels around the atom; this will cause a smearing of the ejected electron pulses. If the resonances have a short lifetime, all of the flux will leave the atom in a short time, and thus few pulses will be ejected. However, the resonances should not be too sharp because then there will be very little electron probability in each pulse. For example, these same states in Li are much sharper than those in Fig. 2, giving many more pulses but very little integrated flux in each pulse.

In Fig. 4 a different energy range is presented. Again the laser polarization is perpendicular to the static field, but now the field strength equals 1.6 kV/cm and the optical pulse duration is ~8 ps. The states may be classified as 19,3,1 at $E = -7.050 \times 10^{-4}$ a.u., 15,9,1 at $E = -6.995 \times 10^{-4}$ a.u., 17,6,1 at $E = -6.947 \times 10^{-4}$ a.u., and 14,11,1 at $E = -6.941 \times 10^{-4}$ a.u. In Fig. 5 the ejected electron flux is plotted as a function of time. Comparing Figs. 3 and 5, notice that the time scale has changed by a factor of 2. The electron pulses are separated by ~30 ps for this case. The difference in energy spacing for these states is ~2% of the energy spacing.

The calculations presented here result in electron pulses of the order of 10 ps in duration. To obtain shorter electron pulses, we can follow two strategies. First, higher static fields (e.g., 10 kV/cm) will result in somewhat shorter electron pulses (typically 1 ps). A second more dramatic improvement can be achieved by applying a two-step laser excitation scheme. The first laser prepares the alkali atom in a stationary Stark state of a low n manifold (e.g., $n = 6$). The wave function of the most blue Stark state (the highest energy state within the n manifold) is strongly localized on the uphill side of the potential. Photoionization by a second laser near threshold of such a blue Stark state is only possible to blue continuum states.[14] Hence, if a short near-infrared laser pulse of ~3 μm (e.g., Ref. 15) is used to photoionize the $n = 6$ blue Stark state, an autoionizing blue wave packet will be excited. Since the energy spacing of blue states of different n manifolds is large, the ejected electron pulses will be short, well below 1 ps.

4. COHERENT CONTROL OF THE EMISSION FREQUENCY

An appealing feature of such a train of electron pulses involves controlling the period of the pulses *while* the pulses are being ejected. The cases we examined for more than two resonances usually involved resonances from different n manifolds. This means that changing the field strength will change the spacing in a complex manner that will tend to cause the wave packet to disperse. However, if the wave packet is constructed from two autoionizing states, the dispersion of the packet is not a problem. For two autoionizing states, the time-dependent flux will have the simple form, $\exp(-\Gamma t)(1 + \cos \omega t)$, if the two states have nearly the same autoionization width Γ and $\omega = 2\pi/\Delta E$. Changing the electric-field strength while the wave packet is on the atom will cause the spacing to change, and thus ω could be controlled during the wave-packet experiment. As an example of this, we discuss two states in Na that were well separated from all other states over the field range of

1.5–1.7 kV/cm. Over this small field range the separation of these two states changed by a factor of 3, which implies a change of electron pulse period by a factor of 3.[16]

In Fig. 6 we present the results of a calculation that shows the real-time tuning of the frequency of ejected electron pulses. For this case we performed a direct numerical integration of Schrödinger's equation using a discrete set of radial basis functions. The excitation of the wave packet was treated as a Gaussian laser pulse that excites states centered at energy, E_0. The equation for the wave-packet part of the pulse was then

$$\left[i \frac{\partial}{\partial t} - H(t) \right] \psi(\mathbf{r}, t) = A(t) x \psi_g(\mathbf{r}), \tag{1}$$

where $A(t) = \exp[-4 \ln(2) t^2 / \tau^2]$, with τ the FWHM of the laser pulse, and the time-dependent Hamiltonian is given by

$$H(t) = H_{\text{atom}} - E_0 + F(t) z. \tag{2}$$

H_{atom} is the static zero-field atomic Hamiltonian, and $F(t)$ is a time-dependent electric field. For the case shown in Fig. 6, $E_0 = -0.00458747$ a.u., $\tau = 8$ ps, and $F(t) = [2.917 \times 10^{-7} + R(t) 2.42 \times 10^{-15}]$ a.u. The function $R(t)$ provides a slow ramp to the electric field. In this calculation $R(t) = 0$ for $t < 30$ ps. This means the initial excitation is performed in a field of 1.5 kV/cm. After 30 ps, $R(t)$ smoothly turns into a linear ramp using the functional form $R(t) = \Delta^4 / (t_c^3 + \Delta^3)$, where $t_c = 30$ ps $= 1.24 \times 10^6$ a.u. and $\Delta = t - t_c$, where both Δ and t_c are in atomic units. At large t, $R(t) \to t - t_c$, giving a linear ramp rate for the field of 0.514 V/(cm ps). The two states that are excited are marked by arrows and are the 23,2,1 state (at $E = -4.641 \times 10^{-4}$ a.u. at $F = 1.5$ kV/cm) and the 22,4,1 state (at $E = -4.535 \times 10^{-4}$ a.u. at $F = 1.5$ kV/cm); their energies at $F = 1.7$ kV/cm are $E = -4.382 \times 10^{-4}$ a.u. and -4.353×10^{-4} a.u.

The spacing of the electron probability pulses in Fig. 6 evolves from ~14 ps near 100 ps to ~40 ps near 400 ps. This is a change in frequency of a factor of 2.5 with mod-

est ramp rates for the electric field. Thus one of the goals of coherent control of atomic processes can be achieved in this system: the experimental control of an atomic scale process through the manipulation of a macroscopic variable. The control of the frequency of the electron probability pulses can be quite accurate since this is simply related to the energy spacing of the two levels.

5. APPLICATION FOR AN ELECTRON GUN

Such a train of electron pulses can, for instance, be used for coherent hammering on surface phonons. We make a coherent electron pulse train, and the repetition frequency of the electric pulses equals the phonon frequency. By sending the train on a surface, the coupling to the phonons will be large if there is a frequency match. Or in other words, the incoming high-energy electron pulse (e.g., 500 eV) will lose a marginal amount of energy to the phonons (few megavolts). The energy loss is within the energy spread of the short electron pulse. It is thus impossible to measure this loss with conventional electron energy loss spectroscopy; however, the time separation of a coherent train of pulses means that only a particular energy can be effectively absorbed by the surface. This technique of coherent hammering is similar to the train of optical pulses that has been used for excitation of optical phonons:[17] the repetition rate of the optical pulse train matches the phonon frequency, while the phonon energy is within the energy bandwidth of an individual optical pulse.

6. CONCLUSION

We have shown that laser-excited alkali atoms in a static field can generate a series of electron pulses. The frequency of the pulses emitted can be controlled through the field strength and the laser frequency that drives the atom from its ground state into the wave-packet state. Furthermore, we have shown that in certain special cases the frequency of the electron pulses can be precisely manipulated *while* the electrons are being emitted. This allows a fine level of control of this system.
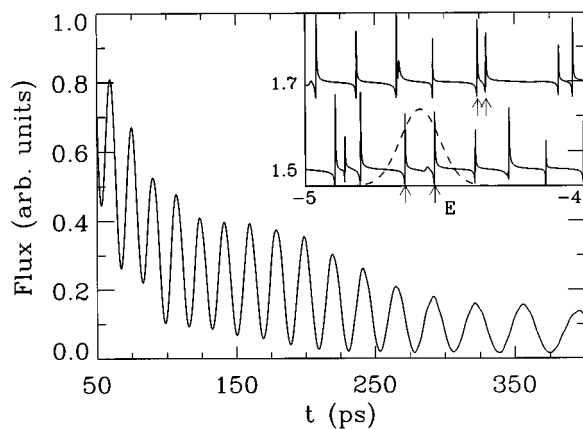
Fig. 6. Outgoing electron flux for a state initially in a 1.5-kV/cm field. The electric field is slowly ramped at a rate of ~0.5 V/(cm ps), which changes the time spacing of the electron pulses from ~14 ps to ~40 ps. The insets show the spectra at 1.5 kV/cm and 1.7 kV/cm (solid curves) and the spectrum of the laser pulse (dashed curve). The energy scale is in 10^{-4} a.u.

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