Designer atoms: Engineering Rydberg atoms using pulsed electric fields

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Rydberg atoms

- one electron excited to a state of large principal quantum number $n$
- physically very large - Bohr radius scales as $n^2$
- weakly bound - binding energy decreases as $1/n^2$

High-$n$ atoms provide a mesoscopic entity that bridges the quantum and classical worlds
Motivation

• explore classical limit of quantum mechanics
• evaluate protocols for controlling and manipulating atomic wavefunctions
• examine concepts for quantum information processing in mesoscopic systems
• examine dephasing and decoherence
• gain insights into physics in the ultra-fast ultra-intense regime
• generate non-dispersive wavepackets

Engineer high $n$ atoms using pulsed electric fields
Engineering Rydberg Wavepackets

Use pulsed unidirectional electric fields of duration $T_p$ with risetimes $T_r \ll T_n$

In limit $T_p \ll T_n$, pulses termed half-cycle pulses (HCPs) and each delivers an impulsive momentum transfer or "kick" to the electron

$$\Delta \vec{p} = - \int F_{\text{HCP}}(t) dt$$

Create desired final state using tailored sequence of HCPs
Realization of Impulsive Regime

Electron orbital period \( T_n = n^3 t_1 \), where \( t_1 = 1.5 \times 10^{-16} \) s

At \( n = 30 \), \( T_n = 4 \times 10^{-12} \) s; \( n = 300 \), \( T_n = 4 \times 10^{-9} \) s

Two approaches:

- use ultra-short (\( T_p < 1 \) ps) freely-propagating HCPs generated by fs-laser-triggered photoconducting switch (Bucksbaum, Jones, Noordam, Stroud)
- use longer HCPs (\( T_p > 500 \) ps) produced by applying output of pulse generator to a nearby electrode

easy to control and measure HCPs, and generate complex HCP trains

Need to work with very-high-\( n \) atoms, \( n > 300 \)
Studies at very high $n$, $n > 300$

Difficult: Rydberg levels closely spaced, atoms strongly perturbed by external fields.

Produce quasi-1D atoms by exciting selected Stark states.
Effect of single HCP: $T_p < < T_n$

Classically: impulse $\Delta p$ changes electron energy by

$$\Delta E = \Delta E_k = \left(\frac{\vec{p}_i + \Delta \vec{p}}{2}\right)^2 - \frac{\vec{p}_i^2}{2} = \frac{\Delta p^2}{2} + \vec{p}_i \cdot \Delta \vec{p} = \frac{\Delta p_z^2}{2} + p_{iz} \Delta p_z$$

Leads to distribution of final $n$ states or, if $\Delta E$ sufficient, to ionization.

Measurements of survival probability used to:

- monitor time evolution of $p_{iz}$
- map distribution of initial $z$-components of electron momentum

Quantum mechanically: impulse(s) produces coherent superposition of states, i.e., a wavepacket

$$|\Psi(t)\rangle = \sum_n e^{-iE_n t} \sum_\ell \langle n\ell m | \Psi(0) | n\ell m \rangle$$

$$\Psi(0) = e^{i\Delta \vec{p} \cdot \vec{r}} |\phi_i\rangle$$

Explore behavior of wavepackets using CTMC simulations
Wavepacket simulations

Employ classical-trajectory Monte Carlo (CTMC) method - Heisenberg time (\( \sim n^4 \)) long - \( \sim 1 \mu s \) at \( n = 300 \)

- initial state represented by appropriate distribution of phase points
- track evolution of each phase point during HCP sequence by solving Hamilton's equation of motion

\[
H(t) = \frac{p^2}{2} - \frac{1}{r} + zF_{\text{train}}(t)
\]

- build up distribution of phase points at time of interest - mirrors probability density distribution of corresponding wavepacket
- consider different times to examine evolution of wavepacket
1D atoms - effect of a single HCP

- induce strong transient phase-space localization
- observed with quasi-1D atoms
- great starting point for further manipulation
Atomic engineering

Use phase-space localized state and tailored HCP sequence to engineer very-high-$n$ ($n \sim 600$) quasi-1D atoms

Apply strong kick in $+z$ direction to localized quasi-1D $n \sim 350$ atoms

Even with pre-localization populate broad distribution of final $n$ states - paradoxically narrow by application of a further HCP
Production of quasi-1D very-high-$n$ states

Apply “elongation” HCP followed by “narrowing” HCP

- observe narrowing of $n$ distribution - counter-intuitive!
- confirmed by SFI measurements
- final quasi-1D state strongly polarized
Physical origin of $n$ focusing

Phase-space portraits of evolution during HCP sequence

- “elongation” kick translates distribution
- product states evolve at different rates
- final HCP aligns with $n \sim 520$
- quasi-stationary final state with narrow $n$ range
Production of quasi-2D near-circular states

- create quasi-1D \( n = 306 \) state oriented along \( x \) axis
- apply dc field step in \( z \) direction - create Stark wavepacket
- turn off field when \( L_y \) maximum

- wavepacket transiently localizes in “Bohr-like” near circular orbit
Wavepacket evolution

Apply dc field of $-20 \text{ mV cm}^{-1}$ for 22 ns - follow subsequent behavior through CTMC simulations

- wavepacket remains localized as “orbits” anti-clockwise
- mimics original Bohr model of atom
- follow time evolution through $\langle x \rangle$, $\langle y \rangle$, and $\langle z \rangle$ and $\langle p_x \rangle$, $\langle p_y \rangle$, and $\langle p_z \rangle$
Wavepacket evolution

- strong oscillations in $\langle p_x \rangle$, $\langle p_z \rangle$ - $90^0$ out of phase
- $\langle p_y \rangle$ ~ constant
- strong oscillations in $\langle x \rangle$ and $\langle z \rangle$ - $90^0$ out of phase
- produce atoms in near-circular “Bohr-like” states
Circular states: experimental verification

Quasi-1D n=306 atoms, dc field of -20 mV cm\(^{-1}\) for 22 ns - probe x and z coordinates of wavepacket with 6ns pulse

- strong oscillations evident
- good agreement with simulations
- produce “Bohr-like” atoms in circular states
- observe periodic changes in momentum distribution
Momentum distribution: time dependence

Examine using 600ps probe HCP applied in z direction

- strong asymmetry observed
- good agreement with simulation
- confirms production of near circular states
- at late times wave-packet dephases
Regeneration of Bohr-like states

- dephasing leads to near-circular states
- apply small in-plane HCP, $\Delta p_0 \sim 0.035$

- gives rise to strong relocalization
- see by probing $x$ and $z$ coordinates of wavepacket
Relocalization: Experimental observations

Probe x and z coordinates with 6ns, 100mV/cm pulse

- strong oscillations seen
- relocalization dependent on kick strength $\Delta p_0$
- good agreement with simulations
- can regenerate at later times with further HCP(s)

$\Delta p_0 = 0.018$
$\Delta p_0 = 0.035$
$\Delta p_0 = 0.10$
1D atoms - effect of multiple periodic HCPs

Impulses all applied in same direction - might expect series of energy transfers leading to ionization

- large fraction of atoms survive
- peak in survival probability seen at $\nu_T \sim 1.3 \nu_n$

Origin of stabilization?

$T_T = 1/\nu_T$

$N = 40 \Delta p_0 = -0.3$
Dynamical stabilization

To survive many HCPs, each must transfer little energy to electron, i.e., require:

$$\Delta E = \Delta p_z^2/2 + p_{iz}\Delta p_z = 0 \Rightarrow p_{iz} = -\Delta p_z/2, \quad p_{fz} = +\Delta p_z/2$$

$p_z$ must then evolve through orbital motion to $-\Delta p_z/2$ at time of next HCP

If electron motion synchronized with HCP frequency obtain dynamical stabilization - see by considering phase space for kicked atom
Phase space for periodically-kicked 1D atom

Poincare surfaces of section

- for $\Delta p < 0$ see islands of stability embedded in chaotic sea
- for $\Delta p > 0$ system globally chaotic
- if initial phase point lies in island remains trapped and survives large number of kicks
- produce non-dispersive wavepacket that undergoes transient phase space localization
- provides opportunity to navigate in phase space
Navigating in phase space

Position of islands depends on kick size and frequency

- steer island away from nucleus by “down chirping” kick frequency

Can control atomic wavepackets using periodic HCP trains - key lies in initial island loading
Selective island loading: CTMC simulations

Take transiently localized state - place at center or periphery of largest island by varying island position, i.e., $T_T$, and $t_d$

- wavepacket circumnavigates island as $N$ increases
- leads to periodic changes in electron energy seen experimentally

Can load localized wavepackets into stable islands
Navigating in phase space: Chirped HCP Train

- load localized $n = 350$ wavepacket into stable island
- down chirp HCP frequency to drive to targeted final $n$ state

\[ \Delta T (N - N+1) = 5.33 + 0.67N \text{ ns} \]

- wavepacket remains trapped
- narrow final $n$ range
- final state strongly polarized
Evolution of $n$ distribution

- monitor using SFI
- as $N$ increases spectra move to higher $n$
- final $n$ distribution narrow, $\Delta n \sim \pm 20$ centered at $n \sim 670$
- by reversing chirp can move to lower $n$
Demonstration of control

Linearly increase $\Delta T$ for 25 HCPs, hold constant for 10 HCPs, linearly decrease for 25 HCPs.

Engineer quasi-1D states of arbitrarily high $n$
Circular states: effect of periodic driving

Produce circular Bohr-like state - try to maintain by periodic driving: $\Delta p_0=0.01$, $T_p=4.3\text{ns}$, monitor with 100ns probe pulse.

- behavior sensitive to frequency and phase of HCP train
- strong oscillations in survival probability seen
- persist while driving continued

Produce non-dispersive wavepacket in near-circular orbit
Periodic driving: Wavepacket evolution

CTMC simulations for driving circular state at orbital frequency

- create a non-dispersive wavepacket that remains localized
- remains locked to driving frequency
- suggests can drive to new \( n \) by “chirping”
Driven circular states: effect of “chirping”

N=100 kicks, $\Delta p_0=0.01$, linear down-chirp from $T_p= 4.3$ to 8.6 ns

- electron motion remains locked to driving frequency
- atom transported to higher $n$ states
- observe using SFI

New opportunity to explore dynamics of driven systems
Wavepacket dephasing

Two causes:

• wavepacket components evolve at different rates - dephases but remains fully coherent enabling revivals - coherent dephasing

• stochastic external perturbations like noise or collisions - leads to irreversible dephasing of wavepacket - decoherent dephasing or decoherence

Decoherence of fundamental importance for all potential carriers of quantum information

Study using a technique that involves electric dipole echoes in Stark wavepackets
Electric dipole echoes

Observe echoes in electric dipole moment of ensemble of Rydberg atoms precessing in an external field after its reversal - analogous to NMR

- produce quasi-1D atoms aligned along x axis
- apply pulsed dc field along z axis to create Stark wavepacket
- monitor wavepacket evolution with probe HCP - see series of quantum beats that map precession of electron orbit

If reverse field at $t = \tau$ observe strong quantum beat echo at $t \approx 2\tau$ - in accord with CTMC simulations
Evolution of Stark states

• classically, electron orbit characterized by energy $E$, angular momentum $L = r \times p$, and Runge-Lenz vector $A = p \times L - \frac{r}{r}$

• in weak field $F$, $L$ and $A$ precess slowly - describe using orbit-averaged values $\langle L \rangle$, $\langle A \rangle$

• define two pseudo-spins $J_\pm = \frac{1}{2} (\langle L \rangle \pm n\langle A \rangle)$ - evolve according to effective Bloch equations

$$\frac{d}{dt} J_\pm = \omega_\pm (F) J_\pm \times \vec{Z}$$

• $J_+$, $J_-$ precess in opposite directions about field

• magnitude of dipole moment varies periodically
Psuedo-spin Precession Frequencies

Hydrogenic energies

\[ E_{n,k,m} = -\frac{1}{2n^2} + \frac{3}{2} nkF - \frac{1}{16} n^4 \left[ 17n^2 - 3k^2 - 9m^2 + 19 \right] F^2 \]

Expressing classical energies in terms of \( J_\pm^z = (m \pm k)/2 \) obtain

\[ \omega_{\pm}(F) = \frac{\partial E(n,J_+^z,J_-^z)}{\partial J_\pm^z} = \pm \left( \omega_k^{(1)}(F) + \omega_k^{(2)}(F) \right) + \omega_m^{(2)}(F) \]

\[ \omega_k^{(1)}(F) = \frac{3}{2} nF \quad \omega_k^{(2)}(F) = \frac{3}{8} kn^4F^2 \quad \omega_m^{(2)}(F) = \frac{9}{8} mn^4F^2 \]

- \( \omega_{\pm} \) depend on \( n \) and F - to first order precession reverses when reverse F
- second-order terms prevent perfect rephasing - minimize using low-\( m,k \) states
- consider behavior in rotating frame
Evolution of Pseudo-Spin Probability Density Distribution

- shown in rotating frame
- consider x,y components \( J_+ \)
- distribution broadens due to dephasing
- pronounced rephasing (echo) following field reversal
- quantify dephasing by considering excess width in azimuthal angle

(a) superposition of extreme parabolic states \( k = n-1, 342 < n < 358 \). (b) initial experimental state
Characterization of Dephasing

Quantify through increases in azimuthal width

- azimuthal width grows linearly in time
- dephasing associated with second-order terms irreversible
- can limit dephasing using periodic reversals
Effect of Periodic Reversals

- field reversed at 100, 300, 500, and 700 ns

- strong quantum beats seen even at late times
- reduced amplitude provides evidence of irreversible dephasing
Noise-Induced Irreversible Dephasing: Decoherence

- colored noise produced by pseudo-random pulse generator
- presence of ±10% amplitude noise damps quantum beats and destroys the echo - introduces irreversible dephasing - decoherence
- can examine effect of the noise frequency spectrum

Stark echoes allow exploration of decoherence in mesoscopic systems on timescales shorter than revivals
Conclusions

- can control and manipulate Rydberg wavepackets with remarkable precision using HCP trains
- Stark quantum beat echoes provide sensitive probe of reversible and irreversible dephasing
- Rydberg atoms form a valuable bridge between the quantum and classical worlds