clude using shaped Rydberg atoms as transient nanostructures for time gated diffraction of x-rays. Another interesting application may be to use sculpted quantum states as registers for quantum computational algorithms. Coherent optical pulses can perform general unitary transformations on quantum states. Coherent radio frequency pulses have already been exploited for quantum algorithms using nuclear magnetic resonance. Shaped optical fields could extend this to electronic states and ultrafast time scales.

This work was performed in collaboration with Thomas C. Weinacht and Jaewook Ahn, and extensive assistance and discussions with them are gratefully acknowledged. Financial support comes from the National Science Foundation under Grant No. 9414335.

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QThA2 **8:30 am**

Reconstructing wavepackets by quantum state holography

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The problem of how to measure the wavefunction of a quantum system in amplitude and phase has attracted a lot of attention over the last few years. In this paper we analyze and further develop our method of Quantum State Holography for reconstructing quantum superposition states in molecules or atoms.[1]

The technique is based on mixing the unknown object state with a known reference state generated in the same system by two delayed laser pulses, and detecting the total time- and frequency-integrated fluorescence as a function of the delay. We show that these data contain enough information to extract the full quantum state of the object wavepacket. The feasibility of the method is demonstrated by numerically simulating the reconstruction of vibrational wavepackets in the $A^1\Sigma_u^+$ potential of sodium dimer. As the test examples we have used (i) a squeezed vibrational wavepacket, and (ii) a vibrational Schrödinger Cat state optically excited by femtosecond laser pulses from the ground $X^1\Sigma_g^+$ electronic potential. Both the cases of completely controlled and noisy relative phase between the laser pulses are considered. In the latter case, we use the technique of Coherence Observation by Interference Noise (COIN)[2] to recover the interference component of the fluorescence signal. Our results clearly demonstrate robustness of quantum state holography, and high quality of wavepacket reconstruction even in the presence of the external noise. Recent related experiments[3] on reconstruction of electronic wavepackets in Rydberg atoms are discussed.

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QThA3 **8:45 am**

Wavepacket dynamics and time-domain spectroscopy in atomic rubidium

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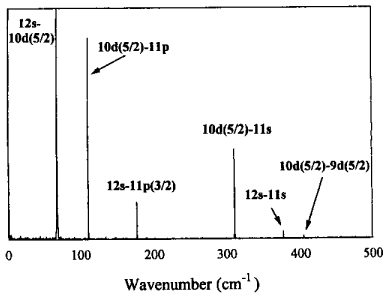
Recently, the detection of atomic wavepackets in alkali vapor by monitoring the coherent UV produced by four-wave mixing was reported.[1] In the pump-probe experiments reported here, the dynamical behavior of wavepackets in Rb has been observed by monitoring the evolving frequency composition of the wavepacket.

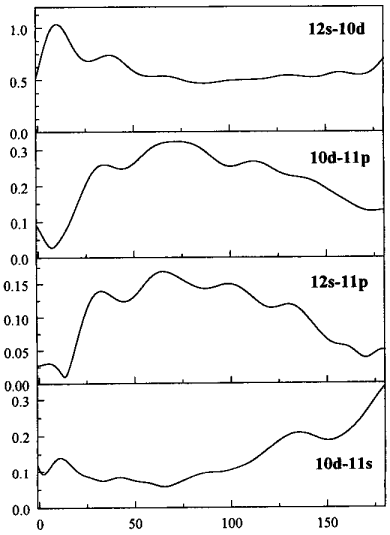
The wavepackets are generated in ~3 Torr of rubidium vapor by 2 photon excitation of the 11s, 12s, 9d, and 10d states with ~100fs pulses centered at 620 nm. Ultraviolet emission is observed near the $np^2P_J \rightarrow 5s^2S_{1/2}$ transitions (n = 7–11), and the spatial and spectral characteristics of the radiation are well described by axially phase-matched four-wave mixing.

The relative intensity of the UV emission associated with a particular ^2P_J fine structure level is monitored as a function of the time delay (Δt) between two, essentially pump and probe, laser pulses. Modulation of the UV signal is observed which is associated with energy level differences between the atomic eigenstates constituting the wavepacket. The time over which beats are observed (Δt ≤200 picoseconds) allows for the frequency components of the wavepacket to be measured to within 0.1 cm^{-1}. Figure 1 shows the Fourier transform of a typical beat signature, and the assigned energy level splittings associated with each peak. Note that the resolution of the experiments allows for frequency components involving the 9d or 10d states to be unambiguously assigned to the J = 5/2 sublevel.

Unexpected behavior of the beat signature has been observed by monitoring the relative amplitude of each beat frequency as a function of Δt. As an example, Fig. 2 illustrates the dependence on Δt of the Fourier amplitudes associated with the 12s–10d, 10d–11p, 12s–11p, and 10d–11s frequency components, while the emission from the $11p^2P_{3/2}$ level was monitored. The general characteristics of each Δt



QThA3 Fig. 1. A Fourier transform of the time-domain signal obtained while monitoring the ultraviolet $11p^2P_{3/2} \rightarrow 5s$ emission from atomic Rb when irradiated by 100 fs pump and probe pulses centered at 620 nm.



QThA3 Fig. 2. The relative amplitudes of the prevalent beat frequencies shown in Fig. 1 are plotted as a function of the delay time between the femtosecond pump and probe pulses.

scan are consistently reproducible, although they do vary depending upon the specific np^2P_J level monitored. Data obtained for the $11p^2P_{3/2}$ transition show that the relative amplitudes of the 10d–11p and 12s–11p components are weak from Δt < 10ps, but grow monotonically and reach their maximum value for Δt ~25–75ps. Notice, too, that d-p and s-p data have essentially the opposite behavior as the s-d components.

To our knowledge, this phenomenon has not been observed previously, and suggests the existence of a time-varying perturbation to the atomic Hamiltonian on the time scale of 10–100 picoseconds. This perturbation is likely to be associated with the interaction of the Rydberg electron with the ion core, and is now being pursued theoretically.

The use of this parametric process for wavepacket detection, as opposed to monitoring ionization or fluorescence, not only provides a conveniently monitored signal, but offers the opportunity to selectively probe the spin-orbit

components of particular Rydberg states. In addition, the results obtained thus far suggest that wavepacket detection in atoms can be used as a real-time probe of the dynamic processes which perturb those atoms. Data obtained by monitoring emission from several np^2P_J states of Rb will be presented and the implications discussed.

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QThA4 (Invited) 9:00 am

Generation and measurement of nonclassical states of molecules

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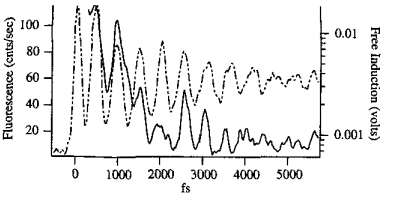
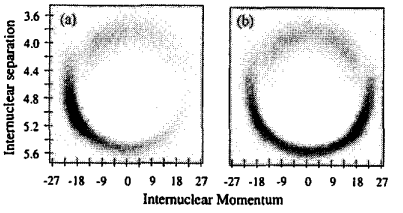
One of the most enticing notions of modern optical physics is the idea that one can manipulate the quantum state of atoms and molecules, and thus "engineer" the properties of matter at its most fundamental level. Homonuclear diatomic molecules are model systems for studying quantum control. They have several degrees of freedom associated with different particles (three are particularly useful: the internuclear vibrational and rotational coordinates and the electronic coordinate) and each degree of freedom has a low dissipation rate as compared to its characteristic frequency. The coupling of the electronic and vibrational degree of freedom allows the preparation of arbitrary vibrational wave packets in both excited and ground electronic states of the molecules by the application of optical radiation.

The first step in the control problem is to determine the form of the (classical) optical pulse sequence that will produce a specific target state of the system. This inverse problem is highly nonlinear, since the final state of the system is a functional of the driving field. As a consequence it is usually necessary to solve the problem using numerical methods. Nonetheless, it is possible to find analytic solutions for the shape of the required optical pulse, even in the regime of complete electronic population transfer from initial to final states, for a wide class of wave packets, under the conditions that (i) the molecule is excited in a time short compared to the vibrational periods of both electronic states and (ii) one selects from the set of possible solutions a pulse with the least rapid turn-on. These constraints ensure that Rabi cycling of the electronic population is averted.[1]

A particularly interesting class of states is one in which several degrees of freedom are entangled. Appropriately shaped pulses cause the excitation of entangled electronic-vibrational states, which may be thought of as consisting of a coherent superposition of a wave packet in the excited electronic state of the molecule and simultaneously a "hole" wave packet in the ground electronic state. Because the characteristic vibrational frequencies of these two states are different, the quantum beats observed in forward scattering are modulated at the commensurate periods of the two wave packets. The suppression



QThA4 Fig. 1. Time-resolved fluorescence (dashed) and free-induction measurements (solid) from wave packets in the A $^1\Sigma_u^+$ and X $^1\Sigma_g^+$ states of K_2. The suppression of the quantum beats at 2000 fs in coherent signal at 830 nm as compared to the fluorescence at 900 nm is consistent with the entanglement of the electronic and vibrational degrees of freedom.



QThA4 Fig. 2. Experimental phase-space quasi-probability densities of the vibrational mode of K_2 in the excited electronic state (A $^1\Sigma_u^+$). Dark areas are those of large positive quasi-probability density, and white negative. The state during (a) the first period and (b) the fourth period after excitation, showing phase-space delocalization.

of incommensurate quantum beats in the forward scattering as compared to the fluorescence at an adjacent wavelength is illustrated in Fig. 1, and we believe this to be a signature of the entanglement.

Several means exist to fully characterize the quantum state of the molecule. The most well established of these, fluorescence tomography, makes use of a projection of the state onto the excited electronic basis.[2] Figure 2 illustrates the results of applying this method to the vibrational component of a potassium molecule in the A $^1\Sigma_u^+$ state. The reconstructed quasi-phase-space density gradually delocalizes over the course of several picoseconds. Since the measurement projects out only the vibrational part of the state, any entanglement of vibrations and rotations appears as dephasing, as does the inhomogeneous distribution of vibrational periods arising from the large number of rotational states occupied initially.

The preparation of arbitrary quantum states and their "readout" is possibly of significance in a variety of endeavours such as demonstrating rudimentary quantum information processing and the control of elementary chemical dynamics.

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QThA5 (Invited) 9:30 am

Application of quantum control theory to manipulate the Zeeman states of an atom

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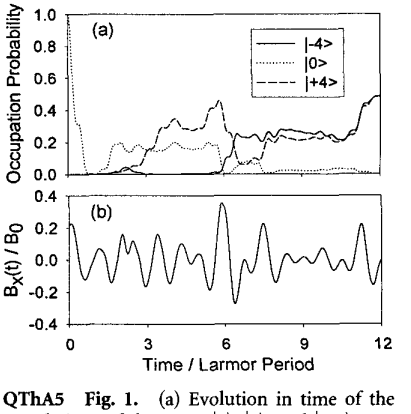
We present experimental results that demonstrate a high degree of control over the spin state of an atom. The experiment is carried out on a sample of about 10^7 laser-cooled ^{133}Cs atoms in their F = 4 ground state hyperfine level released from a magneto-optical trap. Our method is to apply a static magnetic field B_0, a time-dependent transverse magnetic filed $B_x(t)$, and a linearly polarized off-resonance laser field to the atoms. The Hamiltonian is $H = -\mu \cdot \mathbf{B}(t) + M^2\omega_E|M\rangle\langle M|$, where $\mathbf{B}(t) = B_0\mathbf{z} + B_x(t)\mathbf{x}$. B_0 induces a precession of the spins at the Larmor frequency $\omega_L = \mu_B B/4\hbar$. The second term in H arises from the laser field light shift; together with the static field B_0 it yields a finite "anharmonic" ladder of quantum states, labeled by the spin projection quantum number $M = -F, \ldots, F$. Because of the anharmonicity, $B_x(t)$ induces motions of the spin that are not simply rotations of the initial state. Rather, through the application of quantum control theory,[1] it is possible to find a pulse $B_x(t)$ that produces time evolution connecting arbitrary spin states.

We demonstrate this for the particular case of evolution from the state $|0\rangle$ to a "target" state $|\psi_M\rangle = 2^{-1/2}(|4\rangle + |-4\rangle)$, with $\omega_E = 0.02\omega_L$, using the calculated control pulse $B_x(t)$ shown in Fig. 1. We measure the degree to which the target state is reached through a generalization of the Ramsey method of separated oscillatory fields as shown in Fig. 2. We first optically pump the atoms into the state $|0\rangle$, and then apply the control pulse of Fig. 1. We then wait a time T, apply a second control pulse that produces evolution from $|\psi_M\rangle$ back to $|0\rangle$, and finally measure the population returned to the state $|0\rangle$. Figure 3 shows the measured and theoretical population as a function of T. The results show 8 fringes per Larmor period of delay. During the time T, $|\psi_M\rangle$ acquires an additional phase $8\omega_L T$ between its two components, and this causes the state to rephase 8 times per Larmor period. Since only



QThA5 Fig. 1. (a) Evolution in time of the populations of the states $|0\rangle$, $|4\rangle$, and $|-4\rangle$ produced by the control pulse shown in (b).