Information Storage and Retrieval Through Quantum Phase

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Information was stored as quantum phase in an *N*-state Rydberg atom data register. One or more flipped states stored in an eight-state atomic wave packet could be retrieved in a single operation, in agreement with a recent proposal by Grover.

Storage of information as quantum phase was first proposed several years ago in connection with a new class of computational algorithms based on the rules of quantum mechanics rather than classical physics (*1*). The analog of the binary bit, which is the smallest piece of classical information, is the two-level quantum system, or qubit (*2*). Introductions to this subject often begin with the observation that a qubit differs from a bit because it can be prepared in more than two different states (*3*). For example, if the two levels are designated $|0\rangle$ and $|1\rangle$, then superpositions such as $[|0\rangle + \exp(i\phi)|1\rangle]/\sqrt{2}$ are possible states of the qubit. Here, the real number ϕ is the quantum phase difference between probability amplitudes of the two levels in the superposition. Thus quantum phase is present in quantum algorithms, and its maintenance, control, and measurement may be essential factors in the performance of quantum computations.

One example of an algorithm where quantum phase plays an essential role is the database search problem proposed by Grover (*4, 5*). Grover's search differs from some other quantum algorithms because it does not require any nonlocal entanglement of different degrees of freedom in the system. If information is stored as phase, then the superposition principle of quantum mechanics provides an efficient method to search the database (*6*). Superposition is also present in some classical analog data storage media, such as optical holograms, which use the classical phases of interfering electromagnetic waves; however, quantum systems contain the additional nonclassical feature of wave function collapse after a measurement.

In his original paper on this subject, Grover asked the following question: When an *N*-state quantum register (composed of $\log_2 N$ qubits) is prepared with one state phase-shifted from all the others, how many operations does it take to find the flipped state (*4*)? Grover's algorithm takes only order $\sqrt{N}$ steps, whereas classical algorithms require order $N/2$ steps. The algorithm performs an inversion about the average defined by the following unitary operation D on the N-element state vector:

$$D_{ij} = 2/N \text{ if } i \neq j \quad (1)$$

$$D_{ii} = -1 + 2/N \quad (2)$$

This operation amplifies the flipped state and attenuates the others. There have been several recent demonstrations of this search, using qubit systems based on magnetic resonance (*7, 8*) or photon interference (*9*). Grover's second paper considers the case when the database is capable of receiving a single query on all of its states simultaneously. This produces a large payoff in parallelism: The search algorithm now takes only a single quantum operation (*5*).

We have investigated the storage and retrieval of information in the quantum phase of a coherent superposition state of energy levels in a highly excited atom. Two situations were examined. First, we prepared a database in which one of the items was marked. We have demonstrated Grover's contention that the marked item can be retrieved by a quantum computation in a single query of the database, whereas the corresponding information stored in a classical register would require $N/2$ queries on average to find the marked bit. We analyzed the influence of ensemble averaging as well as errors introduced by technical imperfections in our physical system. Second, we can show that the same quantum system and measurement techniques can be extended to store and retrieve large numbers. We have demonstrated storage of numbers up to 2^{N-1} for $N = 8$. A straightforward extrapolation of our results suggests that numbers as large as 2^{100} can be stored in a single N-level atom, where $N = 20$.

In Grover's own words, "the algorithm works by considering a quantum system composed of multiple subsystems; each subsystem has an N-dimensional state space" (*5*). In our experiment, each subsystem consists of N Rydberg states in a single cesium atom, and the quantum system is a collection of these cesium atoms in an effusive atomic beam. Three optical laser pulses intersect this beam. The first one is a 10-ns pulsed laser beam with a line width of $\sim$0.5 cm^{-1}, tuned to 1.08 μm. This beam excites the cesium atoms from the 6s ground states to the 7s state via two-photon absorption. Here we neglect the hyperfine interaction, which is unresolved and has no role in the experiment. This 7s state provides a "reservoir" of probability amplitude that is coherently transferred to the quantum data register.

We used an optically driven unitary transformation, which coupled the subspace of N Rydberg levels to the reservoir, to produce and search the contents of this register. A number of high-lying Rydberg np states are accessible from the 7s state by absorption of a photon from an ultrafast Ti:sapphire chirped-pulse amplified laser pulse (*10*). This contained $\sim$100 cm^{-1} of coherent bandwidth centered at a wavelength of 785 nm. This is the coherent bandwidth contained in a 150-fs pulse, and it can access Rydberg np states in the range $n = 29$ to 39. We used this laser twice in the experiment. The first pulse programmed the quantum register with some information, and the second executed the quantum search for the information.

The optical radiation was programmed with a computer-controlled pulse shaper (*11*), which generates a spectral mask such that N eigenstates were excited. The pulse program also placed information in the quantum register by setting the phase of each quantum state relative to the 7s state. Because the $\langle n\text{p}|z|7\text{s}\rangle$ matrix elements are relatively real, setting the phase is a simple matter of adjusting the relative phase of the optical excitation radiation at each resonant frequency $\omega_{n\text{p}-7\text{s}}$. For example, if $N = 5$ (a five-state register) with principal quantum numbers n_i, $i = 1$ to 5, we can store a binary number 00100 by exciting states $i = 1, 2, 4$, and 5 with a radiation field of the form $\mathbf{E}\cos\omega_{(n_i\text{p}-7\text{s})}t$, whereas state $i = 3$ is excited with radiation $-\mathbf{E}\cos\omega_{(n_3\text{p}-7\text{s})}t$.

A small complication is that different n-states each evolve with a different time dependence $\exp(-i\omega_{n_i}t)$, so that any information stored as quantum phase in the N-state register will also evolve. This effect is just wave packet motion, and we neutralized it by programming the phase of each state to precompensate for phase evolution, so that the phases are easy to interpret at the time they are read out.

In the limit of lowest order perturbation theory, the unitary transformation that transfers population from the 7s state to the N-state Rydberg quantum data register can be represented by a matrix P, whose elements are

$$P_{ii} = 1 \quad (3)$$

$$P_{0i} = P_{i0}{}^* = a_i \exp(i\phi_i),$$

$$a \ll 1 \text{ and } i \geq 1 \quad (4)$$

$$P_{ij} = 0 \text{ for all other } i \text{ and } j \geq 1 \quad (5)$$

The 7s state ($i = 0$) retains most of the proba-

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$$\mathbf{A}\quad A\Psi = \begin{pmatrix} 1 & \varepsilon & -\varepsilon & \cdot & -\varepsilon \\ -\varepsilon & 1 & 0 & \cdot & 0 \\ \varepsilon & 0 & 1 & \cdot & 0 \\ \cdot & \cdot & \cdot & \cdot & \cdot \\ \varepsilon & 0 & 0 & \cdot & 1 \end{pmatrix} \begin{pmatrix} 1 \\ 0 \\ 0 \\ \cdot \\ 0 \end{pmatrix} = \begin{pmatrix} 1 & 7s \\ -\varepsilon & np \\ \varepsilon & (n+1)p \\ \cdot & \cdot \\ \varepsilon & (n+N-1)p \end{pmatrix}$$

$$\mathbf{B}\quad B\Psi = \begin{pmatrix} 1 & \varepsilon & \varepsilon & \cdot & \varepsilon \\ -\varepsilon & 1 & 0 & \cdot & 0 \\ -\varepsilon & 0 & 1 & \cdot & 0 \\ \cdot & \cdot & \cdot & \cdot & \cdot \\ -\varepsilon & 0 & 0 & \cdot & 1 \end{pmatrix} \begin{pmatrix} 1 \\ -\varepsilon \\ \varepsilon \\ \cdot \\ \varepsilon \end{pmatrix} = \begin{pmatrix} 1 \\ -2\varepsilon \\ 0 \\ \cdot \\ 0 \end{pmatrix}$$

Fig. 1. The unitary transformations to (**A**) create the Rydberg data register, and (**B**) retrieve the locations of the flipped bits.

bility amplitude. To match the database search problem of Grover, we make two simplifications: The Rydberg levels are scaled to equal amplitudes, and they are prepared in such a way that at some target time they are relatively real. Information is stored as flipped phase, that is, $\phi_i = \pi$ for some of the states. This preparation of the state is thus represented by $P \rightarrow A$ (Fig. 1A). In this example, the populations of each of the states in the N-state register are equal to $|\varepsilon|^2$, but one of the states is flipped.

At the target time, a second programmed laser pulse with the same amplitudes as in A performs a quantum unitary operation represented by matrix B on these levels by redistributing population through the 7s reservoir state (Fig. 1B). This pulse amplifies the flipped state or states, and suppresses the rest of the states in the wave packet. Although amplitudes are the same as those in pulse A, the phases are all set to π, that is, to the phase used for the flipped states.This pulse is efficient, because it performs its amplification in a single step; and it is universal, because it will decode any binary number stored as quantum phase.

After pulse B, all that remains is to collect the information from the storage medium. The only states that retain population are those whose phases were originally flipped when the information was stored. Each cesium atom is an identical subsystem of the total atomic beam, so we used state-selective ramped-field ionization to identify the states that were amplified in the atoms (*12*). A time-varying uniform electric field F was ramped from zero to several kilovolts per centimeter in 1 μs. The cesium Rydberg np states field ionized according to quantum defect theory at $F \approx [2(n-3.6)]^{-4}$ atomic units of electric field. The ionized electrons were measured using standard particle multipliers, with a detection efficiency near unity. Thus, the value of the electric field at the instant of ionization constituted a measurement of N for the atom. According to the rules of quantum measurement, the probability of finding the atom in state N is proportional to the square of the amplitude for that component of the coherent state. The collection of atoms produced some electrons in each occupied state, thus revealing the binary number that was stored as quantum phase. The minimum number of atoms N_{min} required to measure a single flipped bit depends on the excitation amplitude ϵ defined in Fig. 1. This number was $N_{min} \approx (2\epsilon)^{-2} \approx 100$ for our experimental conditions.

The storage time for the information in the Rydberg quantum register is limited by quantum decoherence, which has several sources. Some of these can be eliminated by improving the physical setup, and some are intrinsic to Rydberg atoms. The Rydberg states survive for several microseconds, which makes ramped-field ionization possible; however, the readout pulse B must arrive within a much shorter time, the radiative lifetime of the reservoir state, which is on the order of several nanoseconds. Our coherence time was also limited by the velocity distribution of the cesium atoms in our laser beam, by black-body radiation, and by Rydberg-Rydberg interactions. We could measure that the coherence time was greater than 1 ns. The ratio of the coherence time for the excited states to the time required to perform an operation on the system is thus at least $O(10^3)$.

In the first series of experiments, we attempted to program and read out a single flipped state in an N-state quantum register. A typical Rydberg register created by the A or B pulse is shown in Fig. 2, A and B, and the measured state populations after amplification with the decoding pulse B are shown in Fig. 2C. All of the states except the one whose phase was flipped were greatly suppressed. Although the experimental protocol outlined above could be carried out in a single measurement, in practice we found some technical difficulty maintaining interferometric stability between pulses A and B. Therefore, we performed 100 identical experiments and used statistical correlation to determine which state was anticorrelated to the others (*13*). In 50 different sets of runs where the stored information was selected at random by a computer, the quantum search method retrieved the correct stored information 100% successfully. We also studied retrieval using fewer trials than 100. To achieve a successful retrieval rate of 95%, we required 13 trials for $N = 6$ and 25 trials for $N = 8$. Of course, this increases the total number of atoms in the measurement to several hundred.

The total number of different values that can be stored as phase in an N-state quantum register is M^{N-1}, where M is the number of different phases that are used. In the simplest case of $M = 2$, we should be able to map 2^{N-1} different numbers into the phase space of N Rydberg states. Retrieval of the information can still be accomplished with a single query of the database, that is, with a single universal unitary operator B. We tested this by loading a computer-generated random number between 1 and 2^{N-1} into pulse A and performing the retrieval experiment a number of times. There were 50 identical trials for each of 50 different randomly

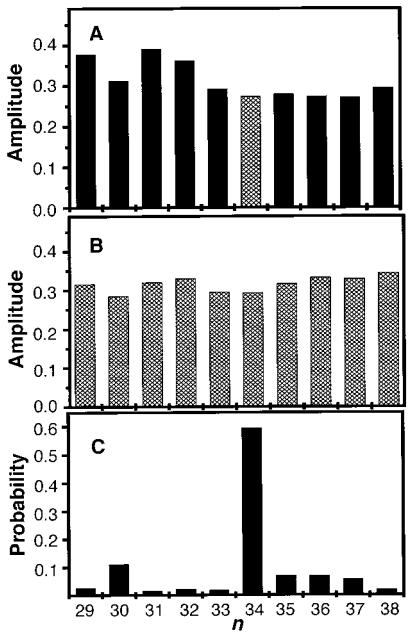

Fig. 2. Rydberg registers made by (**A**) the A-pulse, (**B**) the B-pulse, and (**C**) the $A + B$ pulse. Gray scale represents the phase, which is programmed to have 0 (black) or π (gray) at a target time.

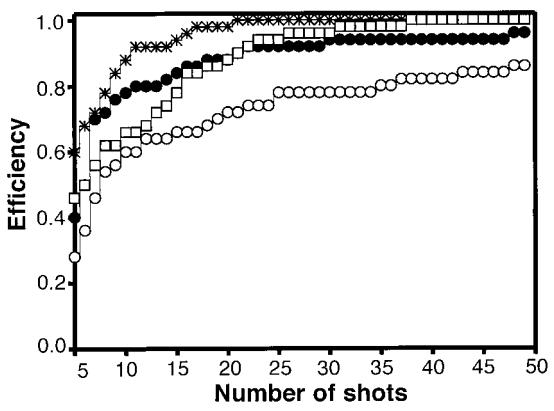

Fig. 3. Results of N-bit coding with six-state quantum register (*) and with eight states (□), and 2^{N-1}-bit coding with six states (●) and with eight states (○). The y axis represents retrieval rate of programmed information extracted from the first M identically prepared pulses, where M represents the number in the x axis.

chosen numbers. We analyzed the $N \times N$ covariance map of electron signals collected from the N Rydberg states to discover which states were flipped. We retrieved the correct number 96% of the time for $N = 6$ and 80% for $N = 8$. These results are summarized in Fig. 3.

One need not confine information storage to $M = 2$ quantum phases per state. In previous work, we demonstrated quantum phase sensitivity for Rydberg wave packets of $\lesssim$10%, which means that more than 30 different numbers can be mapped in a single state (*13*). The maximum number stored in 20 states then reaches $30^{20} \approx 2^{100}$. Other algorithms can be implemented by other unitary transformations such as the application of ultrafast shaped terahertz pulses (*14*). Entanglement of additional degrees of freedom, such as spin and orbital angular momentum, will also extend the reach of this system.

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Mirrorless Lasing from Mesostructured Waveguides Patterned by Soft Lithography

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Mesostructured silica waveguide arrays were fabricated with a combination of acidic sol-gel block copolymer templating chemistry and soft lithography. Waveguiding was enabled by the use of a low–refractive index (1.15) mesoporous silica thin film support. When the mesostructure was doped with the laser dye rhodamine 6G, amplified spontaneous emission was observed with a low pumping threshold of 10 kilowatts per square centimeter, attributed to the mesostructure's ability to prevent aggregation of the dye molecules even at relatively high loadings within the organized high–surface area mesochannels of the waveguides. These highly processible, self-assembling mesostructured host media and claddings may have potential for the fabrication of integrated optical circuits.

Acidic sol-gel self-assembly chemistry (*1–3*) is particularly useful in synthesizing inorganic-organic composite mesoscopically ordered films, fibers, monoliths, and hierarchically ordered structures (*4–9*). Applications of mesoporous materials, for example, as catalysts and separation membranes, have been the main driving force in this field, and their performance in these respects has been extensively documented (*10*). Rather unexplored is the possibility of using mesostructured materials for advanced optical applications. Because of their mesoscopically ordered structures, these materials represent ideal hosts for encapsulating dyes, polymers, and nanocrystals to produce complex multicomponent hybrid materials with interesting and useful optical properties (*11–13*).

We report the fabrication of mesostructured waveguides that exhibit amplified spontaneous emission (ASE), a type of mirrorless lasing (*14*), on low–refractive index mesoporous SiO_2 claddings. A one-step, self-assembly process was achieved by the combination of acidic sol-gel block copolymer templating chemistry with the soft lithographic techniques micromolding, micromolding in capillaries (MIMIC), and microtransfer molding, which have previously been shown to allow the rapid and inexpensive fabrication of liquid-core, polymeric, and inorganic waveguides (*15–17*). In each case, a poly(dimethylsiloxane) (PDMS) elastomeric mold was used to control the shape of the waveguide, whereas block copolymers were used to control the mesostructure.

For waveguiding to occur, the refractive index of the waveguiding medium must be higher than that of its surroundings. Because the mesostructured silica has a refractive index of only 1.43, silicon wafers (refractive index 3.5) coated with mesoporous silica were used as substrates; mesoporous silica is an excellent cladding material because it is mechanically and hydrothermally stable and has a refractive index of only 1.15 (Fig. 5A). The use of these block polymer–derived porous silica materials as low-index-of-refraction silica supports is a convenient route for interfacing the more traditional solid glass, liquid-core, and even polymer waveguides.

To make patterned mesostructures, using MIMIC as an example, we cut open the PDMS stamp at both ends and placed it on a substrate to establish conformal contact. A drop of sol-gel–block copolymer solution (*9, 18*) was then placed at one end of the open microchannels, which were subsequently filled by capillary flow. Gelation of the mesophase precursor solution normally occurred within a few hours. The mold and the resulting mesostructure were left undisturbed for at least 12 hours to allow increased cross-linking and consolidation of the silica network. Then the stamp was peeled off, and the substrate was cleaved to remove the thick film regions on areas that had not been covered by the stamp.

Scanning electron microscope (SEM) images of these patterned mesostructures show line arrays (Fig. 1A) several centimeters long, 1 to 3 μm wide, and 1 to 2 μm high, with 2- to 8-μm spacing. More complex structures with possible optical applications may be fabricated by the use of Fresnel lenses and diffraction gratings as masters to make PDMS molds. These PDMS molds can then be used, for example, to fabricate mesostructured curve or ring patterns (Fig. 1B). The high diversity and complexity of the structures that can be fabricated with the current process offer attractive possibilities for making various optical devices with different functionalities (*9*).

Mesoscopic ordering in the patterned silica-copolymer materials was characterized by low-angle x-ray diffraction (XRD). The

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