Time-domain optical data storage by use of Raman coherent population trapping

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Received September 24, 1993

We experimentally demonstrate time-domain storage and retrieval of amplitude- and phase-encoded optical data, using Raman coherent population trapping, despite the loss of information about absolute optical phases that occurs as a result of the dissipative nature of the process. In this Raman optical storage process homogeneous decay of the optical coherence does not prevent interference between time-separated fields, thus relaxing the requirement for long-lived optical coherences.

There has been much recent interest in time-domain optical storage techniques, especially photon echoes, for information storage[1,2] and processing.[3,4] This is due to the fast response and high storage capacity that can be achieved by use of these techniques with existing cryogenic doped crystals. The frequency- (or time-) domain storage capacities in these materials is given by the ratio of optical inhomogeneous to homogeneous linewidths and can exceed 10^6 bits at a single position.[5,6] However, to satisfy the requirement of long-lived optical coherences, it is necessary to use materials with small optical matrix elements, use highly stable lasers, and operate at liquid-helium temperatures.[1] As a result, practical application of these techniques has so far been limited.

In this Letter we describe a novel technique for storing and retrieving time-domain optical data, using spin coherences excited by resonance Raman coherent population trapping.[7-9] Raman population trapping is an optical pumping process that relies on excited-state decay to create ground-state (spin) coherences. The key property for optical data storage is that these spin coherences are sensitive to the phases of any subsequently applied optical fields.[10,11] This permits optical interference to take place over the entire homogeneous lifetime of the spin coherence, even long after the optical coherence has decayed. Thus constraints on materials for time-domain optical storage are significantly relaxed, requiring that only the ground-state coherences be long lived. Since spin coherences with lifetimes as long as 1 s are known to exist in certain semisolids at room temperature,[12] this technique opens up the possibility of finding high-density optical data storage materials at temperatures beyond the liquid-nitrogen barrier.

Raman echoes employing the coherence between long-lived initial and final states were observed long ago.[13,14] It was also shown that the off-resonance Raman interaction can be made equivalent to direct rf excitation.[15] However, whether Raman-excited spin coherences can be used for optical data storage has not to our knowledge been explored before. The apparent difficulty is that, although these coherences retain the information about the relative phases of the two optical fields used in the Raman excitation process, any knowledge about the absolute phases of these fields is lost because of the dissipative optical pumping processes involved. In this Letter we show that both amplitude- and phase-sensitive optical data storage is possible through Raman population trapping despite this loss of absolute optical phase information.

The resonant Raman interaction is shown in Fig. 1(a). This interaction can be described [see Fig. 1(b)] as optical pumping from the $|+\rangle$ to the transparent $|-\rangle$ state defined by[7-9]

$$|-\rangle = [|a\rangle \exp(i\phi_{12}) - |b\rangle]/\sqrt{2}\,,$$
$$|+\rangle = [|a\rangle \exp(i\phi_{12}) + |b\rangle]/\sqrt{2}\,,$$

where $\phi_{12} = \phi_1 - \phi_2$ is the laser difference phase. Note that the atom in the $|-\rangle$ state contains information about the phase difference between the optical fields but not the absolute phases. On the other hand, an rf $\pi/2$ pulse applied to an atom in state $|a\rangle$ gives

$$\Psi(\pi/2) = [|a\rangle \exp(i\phi_3) - i|b\rangle]/\sqrt{2}\,,$$

where ϕ_3 is the phase of the rf field. If ϕ_3 and ϕ_{12} differ by $\pi/2$ the Raman transparent state and the spin coherence are equivalent.[10,14]

To see how Raman-excited spin coherences store optical temporal information, consider two-pulse exci-

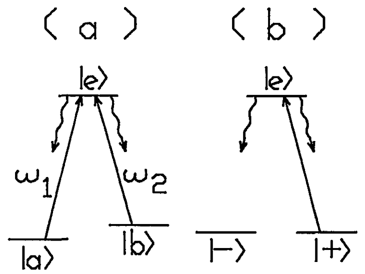

Fig. 1. (a) Schematic of the resonance Raman interaction. (b) The Raman interaction in the coherent state basis. The $|-\rangle$ state is transparent to the resonant optical fields.

tation by Raman resonant optical fields. The pulse separations and durations are assumed long compared with the excited-state lifetime to ensure that the optical pumping model of Raman population trapping is valid. The first pulse creates a population difference between the $|-\rangle$ and $|+\rangle$ states and hence a coherence. However, in the presence of ground-state inhomogeneous broadening, the $|-\rangle$ and $|+\rangle$ states are replaced by

$$\Psi_-(t) = [|a\rangle \exp(-i\Delta t/2) - |b\rangle \exp(+i\Delta t/2)]/\sqrt{2}\,,$$
$$\Psi_+(t) = [|a\rangle \exp(-i\Delta t/2) + |b\rangle \exp(+i\Delta t/2)]/\sqrt{2}\,,$$

where $\phi_{12} = 0$ is assumed and Δ is the laser difference frequency offset of the ground-state transition caused by the inhomogeneous broadening.

When the second Raman resonant pulse arrives at time $t = T$, consider two cases. First, if $\Delta T = 0$ or $n\pi$ (n is an integer), then $\Psi_-(T) = |-\rangle$ and $\Psi_+(T) = |+\rangle$, so that the Raman population trapping induced by this second pulse reinforces the effects of the first pulse (i.e., constructive interference). On the other hand, if $\Delta T = (2n + 1)(\pi/2)$, then Ψ_- has evolved into the $|+\rangle$ state and vice versa, so that the second pulse cancels the population difference (coherence) produced by the first pulse (i.e., destructive interference).

Thus the dissipative process of Raman coherent population trapping permits both constructive and destructive interference between time-separated optical fields and is therefore capable of time-domain optical storage. Of course, some information is lost in the optical decay process, namely, that contained in the absolute optical phase. Hence only time-domain variations that affect the differential optical phase can be stored by Raman population trapping. However, if the optical data are always input as two-frequency Raman resonant pulses, as is the case in our study, the differential optical phase ϕ_{12} contains all the temporal information.

Experimental verification of the ability of Raman coherent population trapping to store and retrieve optical data, spread over times longer than the excited-state lifetime, is accomplished with a sodium atomic beam.[16] The Raman transition used is the $3^2S_{1/2}(F = 1, m = 1) \leftrightarrow 3^2P_{1/2}(F = 2, m = 2) \leftrightarrow 3^2S_{1/2}(F = 2, m = 1)$ at 589.7 nm, with 1.8-GHz ground-state splitting. Ground-state inhomogeneous broadening of 450 kHz is supplied by a magnetic-field gradient. The relevant optical pulse sequence is shown in Fig. 2(a). Actually the laser pulses are supplied by spatially separated cw laser beams intersecting the atomic beam at right angles.[17] Atomic motion along the beam axis translates the indicated spatial distances to time as 145 ns/mm. As shown, the input data consist of two resonance Raman optical pulses, whose separation greatly exceeds the $\Gamma^{-1} = 16$ ns sodium excited-state lifetime. Laser intensities are chosen to give maximum signal-to-noise ratios. We suppress the effects of laser jitter (~1 MHz) by generating all the required optical fields from a single laser, using acousto- and electro-optic modulators.

To recall the stored data, rephasing and probe fields are also required, as shown in Fig. 2(a). To perform rephasing we use an off-resonant Raman excitation[15] rather than direct rf excitation, for experimental simplicity.[18] The Raman resonant optical probe field is needed because the rephased spin coherences do not radiate optically. We accomplish echo detection by monitoring spatial (temporal) variations in probe absorption, in which a spin echo suppresses absorption when it is equivalent to the Raman transparent state.

Figure 2(b) shows the experimentally observed echo signals (demodulated probe absorption versus position), which verify the storage and recall of time-

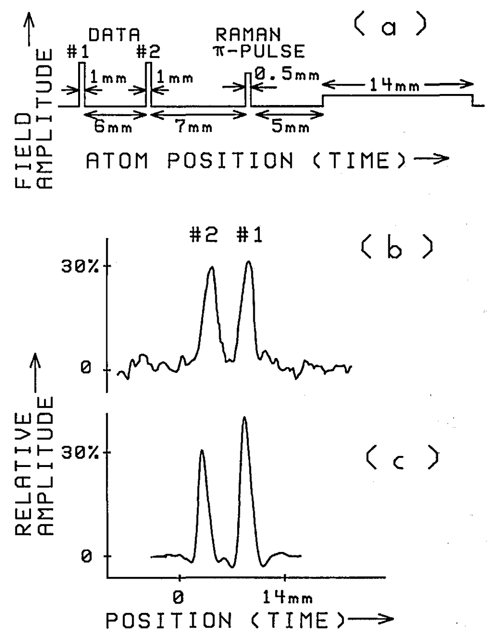

Fig. 2. (a) Experimental pulse sequence for storing and retrieving two-frequency Raman resonant optical data. Pulse widths and separations as indicated. Average laser intensities in units of milliwatts per square centimeter (each frequency): $I_{\text{data\#1}} = 1.2$, $A_{\text{data\#2}} = 0.48$, $I_{\text{probe}} = 0.06$. Off-resonance Raman rephasing pulse: $I_{\pi\text{-pulse}} = 110$ mW/cm^2, detuning +92 MHz. (b) Experimentally observed echo signals demonstrating optical data recall. (c) Corresponding theoretical echoes. Pulse areas in units of $(g^2/\Gamma)\tau$: $A_{\text{data\#1}} = 10$, $A_{\text{data\#2}} = 0.7$. Here $g = g_1/\sqrt{2} = g_2\sqrt{2}$, where g_1 and g_2 are the optical Rabi frequencies and τ is the pulse width. Ground-state inhomogeneous broadening is equal to 400 kHz. An rf π-pulse rephasing field is assumed.

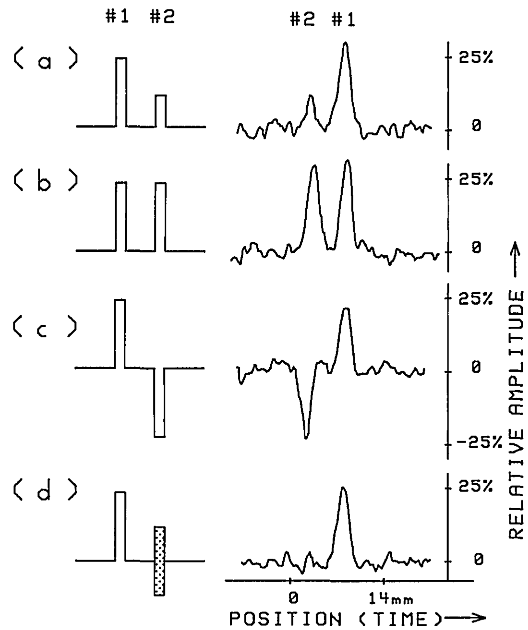

Fig. 3. (a) Echo data when the laser intensity in the second data pulse is reduced by a factor of 2. (b) Echoes for in-phase data pulses [reproduction of Fig. 2(b)]. (c) Echo data when the difference phase of the optical fields in the second data is shifted by π. (d) Echo data corresponding to a $\pi/2$ shift in optical difference phase.

domain optical data by use of Raman population trapping. Figure 2(c) shows the theoretical echo signal calculated by use of a numerical solution of the optical Bloch equations for input parameters that approximately correspond to experimental conditions. We emphasize that these echo signals are spread over time scales that are more than an order of magnitude larger than the optical coherence lifetime, so that the echoes are not produced by optical coherences.

The ability of Raman-excited spin echoes to store and retrieve optical intensity and phase information is illustrated in Fig. 3. Figure 3(a) shows the echo signal obtained under nearly identical conditions to the data of Fig. 2(b), except that the intensity of the second optical data beam is reduced by a factor of 2. As seen, the echo signal reproduces this factor-of-2 attenuation. Figures 3(b)–3(d) demonstrate the differential optical phase sensitivity of Raman echoes. Figure 3(b) is a reproduction of the data in Fig. 2(b), corresponding to identical differential phases in the two optical data beams. In contrast, Fig. 3(c) shows the echo signal when the differential optical phase in the second data beam is shifted by π relative to the first by use of an optical delay line. The negative-going echo signal properly retrieves this phase shift as a sign reversal. For the case of a relative phase shift of $\pi/2$ in the second optical data beam Fig. 3(d) verifies that the second echo signal disappears.

In conclusion, we have shown that, for fast optical decay, Raman-excited spin echoes behave much like conventional spin echoes. The data rate is limited by the inhomogeneous width of the spin transition (in an applied magnetic-field gradient), and the write window is much longer than both the optical homogeneous lifetime and the inverse laser frequency jitter, limited ultimately by the homogeneous spin coherence lifetime. Thus the theoretical time-domain storage density is simply the ratio of inhomogeneous to homogeneous spin linewidth, just as for conventional spin echoes. However, unlike with conventional spin echoes the spatial storage capacity of Raman-excited echoes is determined by the optical wavelength, not by the rf wavelength. Finally, since the Raman spin echo storage process is independent of the absolute optical frequency, it is complementary to, and can be used to enhance, the performance of existing optical spectral-hole-burning memories.

We acknowledge discussions with S. Ezekiel of the Massachusetts Institute of Technology. This study was supported by Rome Laboratory (task 2305D711 and contract F19628-89-K-0030) and by the National Science Foundation (grant ECS-9023746).

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