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OPTICAL SPECTRAL HOLEBURNING WITH RAMAN COHERENT POPULATION TRAPPING

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Abstract Optical spectral holeburning is examined for the case when two-frequency Raman resonant laser excitation is used in place of conventional single frequency excitation. It is found that Raman coherent population trapping can be used to increase storage capacity beyond the limit given by the ratio of optical inhomogeneous to homogeneous linewidths. This is possible because Raman population trapping provides an additional storage dimension. Theoretical analysis and practical issues are presented.

INTRODUCTION

Optical spectral holeburning has generated much recent interest because of its potential for high density optical data storage^{1,2,3} and high speed optical processing.⁴ For example, in rare earth doped crystals the theoretical storage capacity, which is determined by the ratio of optical inhomogeneous to homogeneous linewidths, is typically on the order of 10^6 . Optical data rates or processing speed can be as large the optical inhomogeneous linewidth, which is usually in excess of several Gigahertz.⁵ Although impressive in theory, there are practical limitations which can degrade the storage capacity of rare-earth doped crystals. For example, the storage density is

reduced if there is laser jitter, if the optical data is spectrally broad, or if the operating temperature is well above that of liquid helium.

In this paper, we describe a novel technique for enhancing the storage capacity of optical spectral holeburning materials, especially under adverse conditions. This technique, which is based on Raman coherent population trapping,^{6,7,8} uses simultaneous spectral holeburning on both the optical and ground state transitions. Because ground state holeburning provides an additional storage dimension, the theoretical storage limit is no longer given by the ratio of optical inhomogeneous to homogeneous linewidths alone, but can be larger.

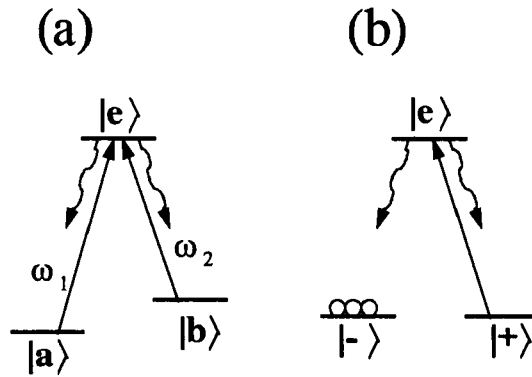


FIGURE 1: (a) Schematic of the resonance Raman interaction. (b) The Raman interaction in the coherent state basis. The $|-\rangle$ state is transparent to the resonant optical fields.

OPTICAL STORAGE WITH RAMAN COHERENT POPULATION TRAPPING

Resonant Raman excitation of a three-level atomic system is illustrated in Figure 1(a). Here, two ground-states $|a\rangle$ and $|b\rangle$ are coupled to a common excited state $|e\rangle$ by resonant laser fields at frequencies ω_1 and ω_2 . For equal optical Rabi frequencies, a non-absorbing state $|-\rangle$ and its complement can be defined as follows,

$$\begin{aligned} |-\rangle &= (|a\rangle \exp(i\phi_{12}) - |b\rangle) / \sqrt{2} \\ |+\rangle &= (|a\rangle \exp(i\phi_{12}) + |b\rangle) / \sqrt{2} \end{aligned}$$

where $\phi_{12} = \phi_1 - \phi_2$, with ϕ_1 and ϕ_2 being the phases of the fields at ω_1 and ω_2 , respectively. Using the $|-\rangle$ and $|+\rangle$ super-position states, the Raman interaction can be re-drawn, as shown in Figure 1(b). As seen, only states $|+\rangle$ and $|e\rangle$ are coupled by the optical fields, and the $|-\rangle$ state is transparent. However, the excited state can decay to either the $|-\rangle$ or $|+\rangle$ state, so that over times longer than the excited state lifetime Γ , the Raman interaction can be modeled as optical pumping into the transparent state (coherent population trapping). For Raman data storage, the important property of the transparent state $|-\rangle$ is that it is equivalent to a ground-state spin coherence.

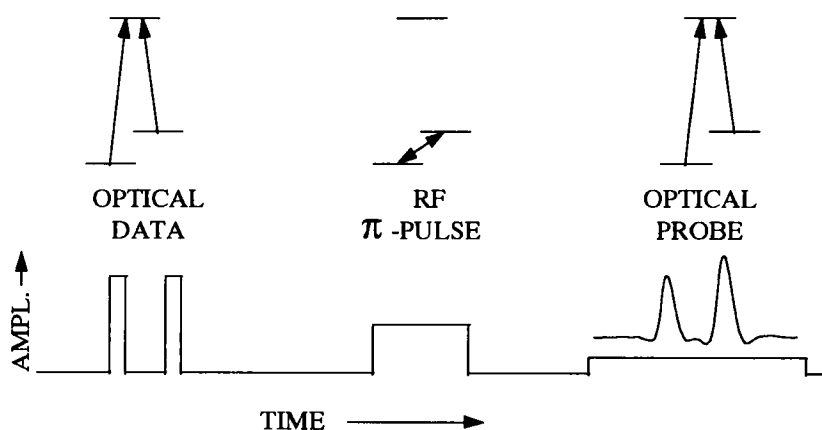


Figure 2: Schematic for optical data storage/recall using Raman spin echoes. Calculated echo signals (probe absorption changes) are displayed above the optical probe pulse.

For the calculation: Data pulse areas in units of $(\Omega^2 / 2\Gamma)\tau$ are: First = 10, Second = 0.7. Here, Ω is the (equal) optical Rabi frequency for each transition and τ is the pulse width. Data pulse separation is $54\Gamma^{-1}$.

To see how coherent population trapping can be used to store optical data, consider the time domain storage scheme of Figure 2. This figure illustrates the storage and recall of two bits of optical data using spin echoes. Both data pulses in the figure contain two Raman resonant optical frequencies. For simplicity, it is assumed that the two optical pulses are separated by much longer than the excited state lifetime. The data pulses create a ground-state spin coherence by optical pumping into the $|-\rangle$ state. For an inhomogeneously broadened ground-state transition, the amplitude of the resulting

spin coherence as a function of frequency stores the time domain information, in analogy to spin echo storage.

To recover the stored information a rephasing pulse is required, which can be rf rather than optical, as shown in Figure 2. If long term storage is desired, the rephasing π -pulse can be replaced by a pair of time separated rf ($\pi/2$)-pulses (i.e. stimulated echoes). Alternatively, it is possible to use optically detuned Raman fields to rephase the spin coherences.⁹ This is more spatially selective than an rf rephasing field, but requires high laser intensities. The resulting spin coherences (echoes) do not radiate optically and hence must be probed. In Figure 2, a low intensity, two-frequency Raman resonant optical probe is used and typical calculated optical absorption signals are as shown. Experimental verification of this calculated result has been accomplished using an atomic beam.¹⁰

Note that in the above example of optical data storage by Raman population trapping, only time-domain information contained in the differential optical phase can be stored. This is because the absolute phase information is lost as the optical coherence decays between data pulses. Next, consider the case when absolute as well as differential optical phase information is stored.

INCREASE OF STORAGE CAPACITY WITH RAMAN POPULATION TRAPPING

Theoretical verification of multiple data set storage and recall, using both Raman and optical time-domain spectral holeburning is accomplished with the pulse sequence shown in Figure 3(a). As shown, the input consists of two time-separated optical data/write pulse sets, where each set consists of a pair of optical data pulses followed by a write pulse. Each data and write pulse contains two Raman resonant optical frequencies. Since the illustrated scheme is similar to a stimulated optical echo scheme, the information in the first data set is stored as a population difference between the $|-\rangle$ and $|+\rangle$ states. But, this population difference is also an rf coherence (since the spin coherences corresponding to the $|-\rangle$ and $|+\rangle$ states are out of phase with each other) and will dephase in the presence of an inhomogeneously broadened ground-state spin transition. Hence, to produce the optical echo, the spin coherence must first be rephased. This is accomplished with the rf π -pulse, as shown. The (two frequency)

optical read pulse is then applied at the appropriate time and a (two frequency) optical echo is produced.

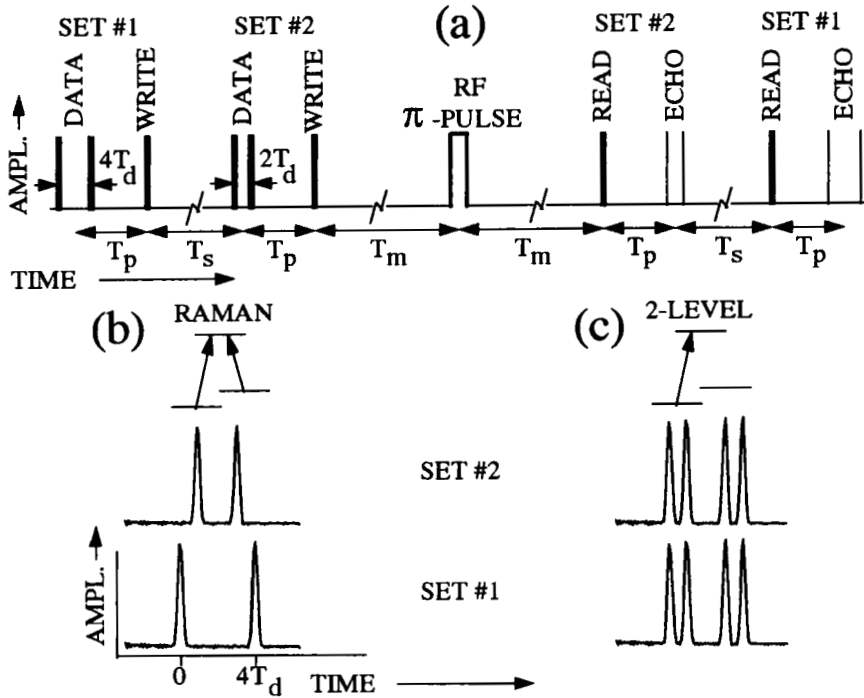


Figure 3: Theoretical demonstration of selective storage of two optical data sets. (a) Pulse sequence used for calculation. Pulse separations (units of excited-state lifetime Γ^{-1}): $T_d = 0.02$, $T_p = 0.2$, $T_s = 20$, $T_m = 14$. All pulse widths are 0.005. Pulse areas: $READ = WRITE = \pi / (10\sqrt{2})$ and $DATA = \pi / (50\sqrt{2})$ for each optical transition. Ground-state inhomogeneous broadening: 5Γ . Optical inhomogeneity: $3 \times 10^4 \Gamma$. (b) Calculated echoes for two-frequency Raman resonant optical pulses, showing selective recall. Note that the output is time-reversed. (c) Echoes for single frequency optical pulses (standard stimulated optical echoes), showing crosstalk.

The fact that the first data set can only be recalled at a particular time after the rf rephasing pulse suggests that it should be possible to write and read additional data sets. To test this, a second data set is applied at later time as shown in Figure 3(a). The resulting calculated echo signals produced by each of the two Raman resonant read pulses is shown in Figure 3(b). As seen, selective recall of both data sets can be achieved. For comparison, Figure 3(c) shows calculated echoes for the case where only

one optical frequency is present in each of the data, write and read pulses. This is equivalent to the standard stimulated optical echo storage scheme. Here, the optical data is stored in the incoherent ground-state populations, which cannot dephase. Thus, selective recall of individual data sets should not be possible, in agreement with the theoretical results shown.

In practice, the highly selective data recall shown in Figure 3(b) may be difficult to achieve. For example, to generate this figure it is assumed that there is no correlation between optical and spin transition frequencies (meaning that each optical spectral hole has full inhomogeneous broadening on the spin transition). Also, it is necessary to have well spaced data sets $T_s, T_m \gg (1/\Gamma) \gg T_p$ and spin inhomogeneous width that is larger than the optical homogeneous width $(1/T_2^{*spin}) > \Gamma$. If these, and other, conditions are not well satisfied, data recall will not be as highly selective (i.e. there will be significant crosstalk).

To overcome the crosstalk problem, preliminary calculations suggest that a non-co-propagating scheme can be used, in analogy those often used in optical stimulated echo experiments. In the case currently under study theoretically, the non-co-propagating data and write pulses have different optical frequencies. For example, the data pulses would be at frequency ω_1 and the write pulse would be at frequency ω_2 , where $\omega_1 - \omega_2$ is Raman resonant. It is not necessary for both optical frequencies to be present simultaneously to generate a spin coherence,¹¹ provided all the pulses arrive within the optical coherence lifetime. The read pulse is chosen to have the same frequency (and direction) as the write pulse, generating an echo pulse with the optical frequency (and direction) of the data pulses. This set of echo pulses should have much lower crosstalk than the co-propagating, two-frequency pulse scheme used to produce Figure 3.

EXPERIMENTAL CONSIDERATIONS

In order to benefit from the additional dimension of storage provided by Raman population trapping, the ground state spin relaxation time T_2^{spin} should be much longer than the optical coherence relaxation time T_2^{opt} . At liquid helium temperature, rare earth

doped crystals generally have nearly equal optical and spin relaxation times and Raman spectral holeburning gives little advantage in theory. A possible exception to this are rare earth dopants in non-magnetic hosts, such as Eu^{3+} or Pr^{3+} in Y_2SiO_5 , where the optical transition is nearly T_1 limited. For example, in $\text{Eu}^{3+}:\text{Y}_2\text{SiO}_5$ with a magnetic field of 100 G, only 1/5 of the optical T_2 is estimated to be due to spin fluctuations,¹² so that up to a factor of 5 increase in storage capacity may be possible.

The greatest advantage to using Raman holeburning arises in applications where it is not possible to take full advantage of the optical transition storage capacity, given by the ratio of optical inhomogeneous to homogeneous widths. For example, in rare earth doped crystals, laser jitter or spectrally broad images like those generated by Rayleigh backscatter from turbulent flow, can span many hole widths. This is expected to decrease the available storage capacity. In such cases, Raman holeburning has the potential to fully restore capacity, since Raman population trapping is not sensitive to the absolute laser frequency, but only the differential frequency.

Another intriguing possibility is to use Raman holeburning to increase the storage capacity of rare earth dopants at high temperatures. For example, at liquid nitrogen temperature $\text{Eu}^{3+}:\text{Y}_2\text{SiO}_5$ has a very short optical relaxation time T_2^{opt} , so that not many optical spectral holes are available. In contrast, spin transitions are expected to broaden much more slowly with temperature, since spin coherence lifetimes in solids T_2^{spin} are on the scale of microseconds even at room temperature.¹³ Thus, Raman holeburning may significantly increase high temperature storage capacity. Of course to give the maximum enhancement in storage capacity, a large inhomogeneous width would be needed on the ground-state spin transition, but this can be supplied with a magnetic field gradient. For Eu^{3+} , the nuclear Zeeman coefficient is about 1 kHz/G, and hence a field gradient of 1 kG/cm over a 1 cm sample would give a 1 MHz inhomogeneous width, which would be adequate for some applications. Alternatively, a rare earth dopant with a larger Zeeman coefficient could be used.

CONCLUSIONS

In summary, we have shown theoretically how Raman coherent population trapping can be used to increase the optical storage capacity of optical spectral holeburning materials

beyond the limit imposed by the ratio of optical inhomogeneous to homogeneous linewidths. This increased storage is based on the additional dimension provided by Raman coherent population trapping.

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