

## Frequency Matching in Light-Storage Spectroscopy of Atomic Raman Transitions

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(Received 18 May 2009; published 28 August 2009)

We investigate the storage of light in an atomic sample with a  $\Lambda$ -type coupling scheme driven by optical fields at variable two-photon detuning. In the presence of electromagnetically induced transparency (EIT), light is stored and retrieved from the sample by dynamically varying the group velocity. It is found that for any two-photon detuning of the input light pulse within the EIT transparency window, the carrier frequency of the retrieved light pulse matches the two-photon resonance frequency with the atomic ground state transition and the control field. This effect which is not based on spectral filtering is investigated both theoretically and experimentally. It can be used for high-speed precision measurements of the two-photon resonance as employed, e.g., in optical magnetometry.

DOI: [10.1103/PhysRevLett.103.093601](https://doi.org/10.1103/PhysRevLett.103.093601)

PACS numbers: 42.50.Gy, 03.65.-w, 06.20.-f, 07.55.Ge

Optically dense atomic media can be made transparent by means of destructive quantum interference of optical absorption amplitudes [1]. Media prepared under such conditions called electromagnetically induced transparency (EIT) are known to show interesting properties, such as extremely slow group velocities [2–4]. Storage of light can be achieved by adiabatically reducing the group velocity to zero allowing for reversible storage of photonic information in an atomic ground state coherence. In recent experiments also nonclassical light states such as squeezed and single-photon states have been reversibly stored in atomic media [5–8]. Light storage promises to be of importance in the field of quantum information, where applications as the implementation of quantum storage devices, gates, and generation of photonic qubits have been suggested [9–11]. Electromagnetically induced transparency has been shown to enable metrological applications like optical magnetometry. Because of the narrow linewidths obtainable with dark resonances [12], precise magnetometers can be realized [13,14]. Recent advances in this field have led to the development of state-of-the-art magnetometers surpassing the precision of superconducting quantum interference devices [15]. Recently, it has been demonstrated by some of the authors that the storage of light in an atomic tripod system after retrieval allows for the observation of an optical beat signal resonant to an atomic ground state coherence [16].

Here, we demonstrate that the observed resonance beating is a general feature of light storage in optically dense ensembles of atoms with dark states. Storage of light was performed by dynamically reducing the optical group velocity in the dark-state system to zero. After reaccelerating the stored light, a beating between the released signal beam pulse and a control beam field is observed which is shown to match the energetic splitting between the two ground state sublevels for any two-photon detuning of the input

pulse within the EIT transparency window. The frequency matching effect is explained in terms of the polariton picture of EIT [17,18].

The observed synchronization effect allows for a novel method for atomic spectroscopy between ground state sublevels, as is of interest both in the field of atomic clocks and the measurements of magnetic fields. Our experimental measurements indicate that with light storage spectroscopy, two-photon resonance frequencies can be measured within a fraction of the time required for the acquisition of a complete dark resonance spectrum. Furthermore, the scheme is relatively robust against fluctuations of the precise optical frequencies of the two incident light fields. As long as difference frequency fluctuations do not exceed the spectral width of the dark resonance, the retrieved frequency difference remains locked to the two-photon resonance.

Consider a medium consisting of atoms exhibiting a level structure with two stable ground states  $|g_{-}\rangle$ ,  $|g_{+}\rangle$  and one spontaneously decaying electronically excited state  $|e\rangle$ , as depicted in Fig. 1. The transition between the states  $|g_{+}\rangle$  and  $|e\rangle$  is driven by a  $\sigma^{-}$ -polarized “control” beam, which is assumed to be stronger than the  $\sigma^{+}$ -polarized “signal” field coupling the states  $|g_{-}\rangle$  and  $|e\rangle$ . Under the conditions of electromagnetically induced transparency, the propagation of light can be described in terms of a moving quasiparticle, the so-called dark-state polariton, which is a mixture of a photonic contribution and an atomic spin-wave component [17]. For an ensemble of three-level atoms, as shown in Fig. 1, interacting with a strong control field and a weaker signal field, the group velocity is proportional to the intensity of the control field, and thus can be manipulated experimentally. The atom-light quasiparticle even can be stopped by dynamically reducing the optical field intensities to zero, which is referred to as “storage of light” [6,7]. During this process,

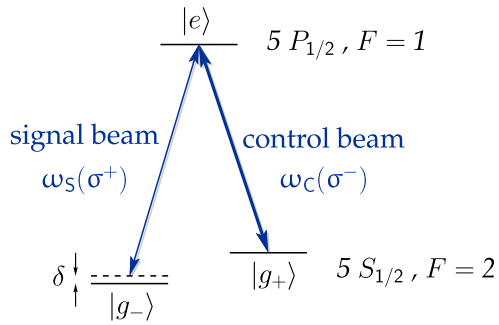


FIG. 1 (color online). Three-level scheme used for storage and retrieval of light. The levels  $|g_{+}\rangle$  and  $|e\rangle$  and  $|g_{-}\rangle$  and  $|e\rangle$  are coupled by a  $\sigma^{-}$ -polarized control field and a  $\sigma^{+}$ -polarized signal field with optical frequencies  $\omega_c$  and  $\omega_s$ , respectively. The two-photon detuning is given by  $\delta = \omega_s - \omega_c - 2g_F\mu_B B$ , where  $\mu_B$  is the Bohr magneton,  $g_F$  the hyperfine  $g$  factor, and  $B$  the magnetic field.

information about the polarization state, phase, and amplitude of the incident light field is reversibly mapped onto a spin-wave coherence. After reactivation of the control beam, these properties are again imprinted on the reaccelerated signal field. It is remarkable that pure quantum states of light can in this way be reversibly stored in the multiparticle system of an atomic gas [19].

The apparatus used in our experiment (see Fig. 2) is a modified version of a previously described setup [16,20]. A grating stabilized diode laser locked to the  $F = 2 \rightarrow F' = 1$  component of the rubidium  $D1$ -line near 795 nm is used as a laser source of both the optical signal as well as the control field. The two beams pass independent acousto-optical modulators to allow for a variation of the difference frequency and intensity of the individual beams. Inside a magnetically shielded region, a 50 mm long rubidium vapor cell containing 10 torr neon buffer gas is heated to approximately 80 °C. A magnetic bias field directed along the optical beam axis is applied in order to split up the ground state Zeeman components, which serve as states  $|g_{-}\rangle$  and  $|g_{+}\rangle$  in the experiment. The energetic separation of two neighboring ground states is given by  $g_F\mu_B B$ , where  $\mu_B$  is the Bohr magneton, and the hyperfine  $g$ -factor equals  $g_F = 1/2$  for the used transition.

After passing the acousto-optic modulators, the beam paths are spatially overlapped and sent through a polarization maintaining optical fiber. The beams are then expanded to 2 mm beam diameter and sent through the

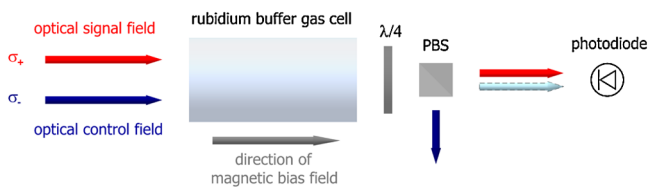


FIG. 2 (color online). Scheme of experimental setup.

rubidium cell with opposite circular polarizations. In our experiments, the control beam intensity was always much stronger than the signal beam so that most of the relevant atoms were in the  $F = 2, m_F = -2$  ground state sublevel. In this case, the states  $|g_{-}\rangle$  and  $|g_{+}\rangle$  of the simplified three-level scheme shown in Fig. 1 correspond to the  $m_F = -2$  and  $m_F = 0$  Zeeman sublevels of the  $F = 2$  ground state component, respectively. Under EIT conditions, a dark coherent superposition of these ground states is established by optical pumping. After traversing the rubidium cell, with a  $\lambda/4$ -wave plate, the circular polarizations of signal and control beams can be converted to opposite linear polarizations, respectively, and a subsequent polarizing beam splitter removes the control beam, so that only the signal beam intensity is detected with an optical photodiode. In order to observe an optical beating signal between control and signal fields, a small portion of the control field is required to reach the detector, which can be realized by a slight rotation of the  $\lambda/4$  plate.

In initial experiments, we investigated both dark resonances and the storage of light in our setup. Typical values for the optical field power were 300  $\mu\text{W}$  for the control and 100  $\mu\text{W}$  for the signal beam. Subsequently, the presence of a beating signal was verified by irradiating the sample with initially rectangularly shaped optical signal field pulses with a duration of approximately 60  $\mu\text{s}$ , with its falling edge set to the same time as the falling edge of the control field. After a storage period of typically 20  $\mu\text{s}$ , only the control field was turned on again triggering the retrieval of the signal field. The detected signal, as shown in Fig. 3(a), showed rapid oscillations on top of an exponentially decaying amplitude signal due to the beat between the signal and control fields. The oscillation frequency between the control field and the initial or restored signal field was determined by fitting the recorded signal with a sinusoidally modulated function.

We have recorded such oscillation data for different values of the magnetic bias field, while the values of the optical frequencies of signal and control fields were left constant. Figure 3(b) shows the measured value of the beat frequency between signal and control field as a function of the bias field. The data points can be well fitted with a linear function, as shown by the dashed line. Within our experimental accuracy, the beat frequency between the released signal light and the control beam frequency corresponds to the frequency of the  $\Delta m_F = 2$  transition between states  $|g_{-}\rangle$  and  $|g_{+}\rangle$ . That is to say, provided that one remains within the dark resonance window, storage can be done with a frequency difference somewhat differing from the precise two-photon resonance with  $\delta = 0$  (where  $\delta = \omega_s - \omega_c - 2g_F\mu_B B$  denotes the two-photon detuning), the released signal beam frequency is such that the optical difference frequency then matches the two-photon resonance. In additional measurements, we have performed the storage procedure with a variable difference frequency,

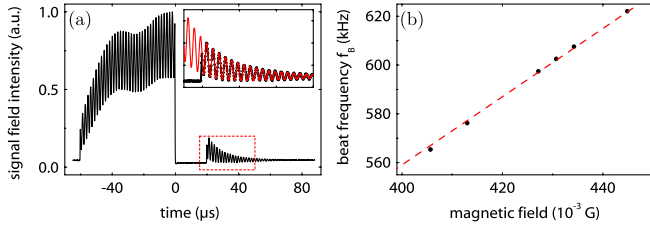


FIG. 3 (color online). (a) Typical stored light signal for a storage period of  $20 \mu\text{s}$ . At time  $t = 0$ , the control field and the signal field were switched off adiabatically, and at time  $t \approx 20 \mu\text{s}$ , only the control field was switched on, restoring the signal field. The inset shows a portion of the retrieved pulse fitted with a sinusoidally modulated function [22]. (b) Measured beat frequency of the released signal light as a function of the applied magnetic bias field. All data points were recorded with the same value of the two-photon detuning of the initial signal field. The size of the error bars of the data points is smaller than the drawing size of the dots.

while the magnetic bias field was left constant. Figure 4 shows a measurement of the observed beat frequency between the released signal beam and the control beam. The shown circles are the corresponding data points, which have been fitted with a linear function (solid line). The slope of the fitted line is  $-0.02(\pm 0.02)$ , which within our experimental uncertainty is consistent with a slope of zero. For detuning values comparable to the width of the EIT window, the amplitude of the released signal is strongly damped, resulting in a diminished signal-to-noise ratio. For comparison, the data points shown as squares, which have been fitted with a linear function shown as a dashed line, give the measured beat signal of the signal and control beams before storage.

We now show that the matching effect of the two-photon resonance upon retrieval can be explained within the dark-

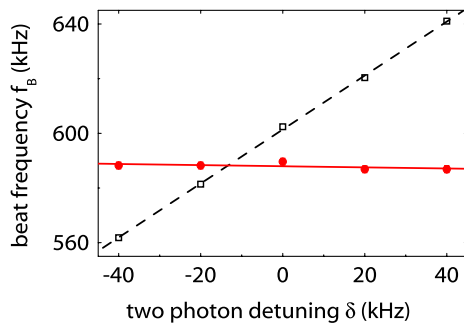


FIG. 4 (color online). Beat frequencies for different values of the two-photon detuning for the pulses before storage (open squares) and after the retrieval procedure (circles). For all shown data points, the atomic sample was exposed to the same magnetic bias field. The slopes of the fitted linear curves are (within uncertainties calculated by the fitting routine) one for the input pulses (dashed line) and zero for the pulses detected after storage (solid line). The size of the error bars is within the drawing size of the data points.

state polariton picture of EIT [17]. For simplicity, we assume that the initial two-photon detuning is small so that the following condition is satisfied [18]:

$$\delta \ll \frac{g\sqrt{N}}{\sqrt{\Gamma T}}. \quad (1)$$

Here,  $\Gamma$  is the upper state relaxation rate,  $g$  is the signal field coupling strength,  $N$  is the density of atoms, and  $T$  is the time that the signal pulse propagates inside the medium before the control field is completely turned off. Under this condition, the absorption of the signal field can be neglected. Introducing normalized, temporally, and spatially slowly varying amplitudes of the complex signal field, as well as the control-field Rabi frequency,  $E(z, t) = \sqrt{\hbar\omega_s/2\epsilon_0}\mathcal{E}(z, t)e^{-i\omega_s(t-z/c)}$ , and  $\tilde{\Omega}(z, t) = \Omega(t - z \cos\alpha/c)e^{-i\omega_c(t-z \cos\alpha/c)}$ , where  $\omega_s$  and  $\omega_c$  are the carrier frequencies of signal and control fields, and  $\alpha$  is the angle between the propagation axes of signal and control fields, the dark-state polariton reads

$$\Psi(z, t) = (\cos\theta)\mathcal{E}(z, t) - (\sin\theta)\sqrt{N}\rho_{g_+g_-}(z, t). \quad (2)$$

Here,  $\rho_{g_+g_-}(z, t)$  is the temporally and spatially slowly varying density matrix element in the two ground states  $|g_+\rangle$  and  $|g_-\rangle$ .  $\theta = \theta(t - z \cos\alpha/c)$  is the mixing angle determined by the amplitude of the control field via  $\tan\theta = g\sqrt{N}/\Omega(z, t)$ , where  $\Omega(z, t) = \Omega(t - z \cos\alpha/c)$ . It is important at this point to keep the retardation of the control field. As has been shown in Ref. [18], if the two-photon detuning  $\delta = \omega_s - \omega_c - \omega_{g_+g_-}$ ,  $\omega_{g_+g_-}$  being the two-photon resonance frequency, is sufficiently small, the propagation equation of the dark-state polaritons reads

$$\left(\frac{\partial}{\partial t} + c\cos^2\theta(z, t)\frac{\partial}{\partial z} - i\delta\sin^2\theta(z, t)\right)\Psi(z, t) = 0. \quad (3)$$

Solving this equation, one finds that an input signal pulse with two-photon detuning  $\delta_0 = \omega_s^0 - \omega_c - \omega_{g_+g_-}$  is mapped onto a stationary spin wave with a phase oscillating in  $z$  direction with  $(\omega_s^0 - \omega_c)(1 - \cos\alpha)/c$ . Note that if both fields copropagate, i.e., if  $\cos\alpha = 1$ , the spatial phase of the stored spin wave actually vanishes. This is because the front of the control pulse which switches the moving polariton to a stationary spin wave propagates with the vacuum speed of light and thus moves in phase with the beat note between control and signal. Upon retrieval of the pulse with a control field corresponding to a mixing angle  $\cos^2\theta_r$ , the polariton oscillates at a frequency equivalent to a two-photon detuning  $\delta$  given by

$$\delta = \delta_0(1 - \cos\alpha)\cos^2\theta_r, \quad (4)$$

where it was assumed that the direction of the control field was not changed. One recognizes that for copropagating control and signal pulses, i.e., for  $\cos\alpha = 1$ , the two-photon detuning of the retrieved pulse vanishes exactly irrespective of its input value as long as the latter lies

within the EIT transparency window. If control and signal beam propagate in orthogonal directions, the frequency of the retrieved signal pulse is pulled towards two-photon resonance with the strength of pulling given by the ratio of the group velocity at read out to the vacuum speed of light. The latter can be understood as follows: If the storage generates a spin wave with nonvanishing  $\vec{k}_{\text{spin}}$ , i.e., for  $\cos\alpha \neq 1$ , phase matching demands that the momentum of the retrieved probe field fulfills  $\vec{k} = \vec{k}_{\text{spin}} + \vec{k}_c$ , where  $\vec{k}_c$  is the  $k$  vector of the control field at readout. If the group velocity at read out is not equal to  $c$ , i.e., if  $\cos^2\theta_r \neq 1$ , the medium has however an index of refraction which deviates slightly from unity for a small two-photon detuning. The latter is responsible for the factor  $\cos^2\theta_r$  in Eq. (4).

To conclude, we have shown that light stored in  $\Lambda$ -type three-level systems shows a strong pulling of its carrier frequency towards two-photon resonance upon retrieval. This effect which holds for an arbitrary two-photon detuning of the input field within the EIT transparency window is a nondissipative effect and can be used to measure the energetic separation of the ground states without precise knowledge of the input signal frequency. Within our experimental accuracy, the beat frequency between the retrieved signal field and the control field does not depend on the initial two-photon detuning of the pulses before storage. Since the lower levels of the  $\Lambda$  system are in our case Zeeman sublevels of the Rb ground state, the beat frequency between the released signal and the control beam is expected to resemble  $2g_F\mu_B B$ , where  $g_F$  denotes the hyperfine  $g$  factor that equals  $1/2$  for our rubidium system. The used method represents a novel approach for optical (Raman) spectroscopy that should also be applicable to two-photon transitions with a ladder level scheme. In preliminary experiments, the obtained accuracy of a magnetic field measurement carried out by recording the spectrum of a dark resonance was also achieved by evaluating the signal generated by a single retrieved signal light pulse, but with a greatly reduced measurement time. We wish to point out that our light storage spectroscopy should also be applicable to nonclassical light states [5,21]. Since the light storage, despite the frequency conversion, is expected to preserve the quantum properties of light, when squeezed light is stored, beat frequency measurements of the released light should be possible with sub-shot-noise precision. This opens up possibilities for novel quantum limited measurements of atomic transition frequencies and magnetic fields.

G.N. acknowledges support by the Alexander von Humboldt Foundation.

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- [22] The shape of the transmitted (initially rectangular shaped) input pulse is due to absorption in the dense medium. Upon entry into the cell, the atoms are pumped into the dark state before the pulse can propagate lossless. The fast oscillations on the input signal are due to beating of the incident optical fields. The shape of the retrieved signal light is determined by the Lorentzian-like spectrum of the transparency window, whose Fourier transform is an exponentially decaying function.