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Coherent population trapping in a crystalline solid at room temperature

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Observation of coherent population trapping (CPT) at ground-state Zeeman sublevels of Cr^{3+} ion in ruby at room temperature is reported. A mechanism of CPT, not owing to optical pumping, is revealed in a situation when the optical pulse duration is shorter than the population decay time from the excited optical state.

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In the last few decades much attention has been paid to studying optical interference phenomena originating from the atomic coherence excited by laser light while interacting with multilevel atomic media. These phenomena include coherent population trapping (CPT) [1], electromagnetically induced transparency (EIT) [2], lasing without population inversion (LWI) [3], slow light [4], and many others. The interest to this field arises from a number of potential applications of these phenomena. Among them are quantum information storage and processing, ultrasensitive magnetometry, metrology, the development of lasers in vacuum ultraviolet (VUV), x-ray and γ-ray ranges, enhanced nonlinear optical conversions, etc. CPT and related phenomena are being widely studied in gaseous media, mostly at Zeeman or hyperfine transitions of alkali metal vapors. However, from the practical viewpoint solid-state materials are preferable. The advantages of solids are high atomic density, absence of atomic diffusion, compactness, and robustness. At the same time, decoherence processes in solid media are severe. Nevertheless, a few demonstrations of EIT and slow light in $Pr^{3+}:Y_2SiO_5$ and nitrogen-vacancy centers in diamond [5,6] and nondegenerate four-wave mixing [7] in solid hydrogen have been reported by several groups. However, all those works were performed at temperatures close to the liquid helium one. This fact, in turn, makes real solid-state applications of the above phenomena questionable. Moreover, low-temperature experiments cited above EIT were observed only for a small portion of ions within the inhomogeneous linewidth of the optical transition (typically, 0.01–0.1% of all ions within the laser jitter were involved). The latter diminishes the most important advantage of solids—their high density. However, most optical transitions in doped solids are homogeneously broadened at room temperature. Thus, it is possible to make all dopant ions participate in CPT and EIT.

In gaseous media, such as alkali-metal vapors, CPT occurs due to the optical pumping of atoms into a coherent superposition of ground-state sublevels, which does not interact with the incident laser radiation (the so-called "dark" state). This mechanism works well if the population decay is much faster (typical lifetimes are tens of nanoseconds) than decoherence processes at the transition between the ground-state sublevels. (Decoherence time can be made as long as several seconds.) The situation is completely the opposite for the case of solids, where decoherence processes are much faster than any population relaxations. The two rates can be made comparable and the usual mechanism of CPT, related to optical pumping into the dark state, can be efficient only at very low temperature.

In this Rapid Communication I illustrate theoretically and report on experimental testing of transient CPT based on a slower rate of optical excitation in the presence of ground-state coherence in a situation when the duration of an optical pulse or a pulse train is much shorter than the excited state population relaxation. On the basis of the proposed pulsed excitation of the ground-state coherence, I report an experimental observation of CPT in a crystalline solid at room temperature.

First of all, we have to identify the class of solid-state materials in which the phenomena of EIT and CPT can be observed at room temperature. I will consider only optical crystals doped with either rare-earth or transition metal ions possessing discrete electronic states within the band gap of the host material. The class of ions, in which electron paramagnetic resonance (EPR) can be observed at room temperature, is of particular interest. It includes Cr^{3+}, Eu^{2+}, Gd^{3+}, and several others. The most important feature of those ions is that, when incorporated into a crystal, their first excited electronic state lies several thousands cm^{-1} above the ground one. Consequently, the strongest relaxation process between the ground-state Zeeman or hyperfine sublevels, owing to resonant inelastic scattering of phonons at low-energy electronic transition (Orbach relaxation, [8]), is suppressed. As a result, the room temperature decoherence rate at the transitions between Zeeman or hyperfine sublevels can be as low as a few MHz. Some of those ions, namely, Cr^{3+}, Eu^{2+}, and Gd^{3+}, offer very favorable optical properties as well. In the following discussion, the case of ruby, i.e., $Cr^{3+}:Al_2O_3$, is considered.

In order to see how CPT can be observed under pulsed excitation, let us consider the following illustrative theoretical model. A three-level medium is illuminated with a resonant laser pulse interacting with both $1 \leftrightarrow 3$ and $2 \leftrightarrow 3$ optical transitions (see the inset of Fig. 1). For the rest of the paper we will be interested in the amount of population transferred into level 3 by a laser pulse or a pulse train as a function of the energy separation between ground-state Zeeman sublevels 1 and 2 which can be varied by means of an external magnetic field. The following notations are introduced: Γ is the homogeneous half-width of optical transitions, γ is the Zeeman decoherence rate, and Ω is the laser field time-dependent Rabi frequency (dipole matrix elements of both optical transitions are assumed equal). The initial populations of the two ground-state sublevels are $\rho_{11}=\rho_{22}=1/2$, $\rho_{33}=0$. The symmetry of the system implies that populations of the ground-state sublevels are always equal. Assuming the pulse

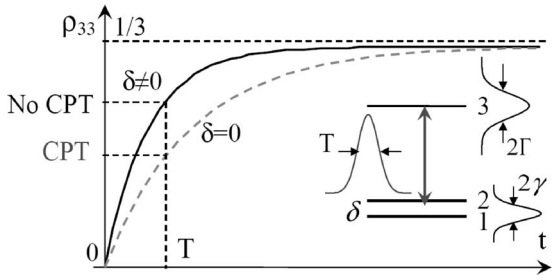

FIG. 1. Temporal evolution of the upper optical state population under pulsed excitation according to Eq. (1). The inset shows the model of a three-level atomic medium interacting with a light pulse.

duration is longer than Γ^{-1}, one can adiabatically eliminate optical polarizations. The assumption that pulse duration is longer than γ^{-1}, relevant under the experimental conditions described below, allows one to algebraically solve the density matrix equation for Zeeman coherence as well and to remain with only one equation for the upper state population:

$$\frac{d\rho_{33}}{dt} = P\left[1 - \frac{P(P+\gamma)}{\delta^2 + (P+\gamma)^2}\right](1-3\rho_{33}), \qquad (1)$$

where $P=2\Omega^2/\Gamma$ and δ is the frequency separation between the ground-state sublevels. The second term in brackets represents the effect of Zeeman coherence on the rate at which level 3 is populated. The laser pulse starts acting on the medium at $t=0$ and level 3 gets populated as shown in Fig. 1. The growth rates are different for $\delta=0$ and $\delta\neq 0$. If at some instant T the laser pulse is shut off, the final population of level 3 is smaller for $\delta=0$ than it is for $\delta\neq 0$ because of its lower growth rate as illustrated in Fig. 1. However, as $t\rightarrow\infty$, all three levels become equally populated, as expected. In other words, Zeeman coherence slows down the excitation of the upper optical state though its population at $t\rightarrow\infty$ is the same for $\delta=0$ and $\delta\neq 0$. Thus, in this situation CPT has transient nature and can occur only for finite pulse duration. The width of this transient CPT resonance is determined by $P+\gamma$, i.e., by the Zeeman decoherence rate and power broadening.

Similar effects can be observed under the illumination of the medium with a short (nanosecond) train of ultrashort (picosecond) pulses. In addition to CPT resonance at a zero magnetic field, one should expect drops in the upper state population as the Zeeman sublevel separation becomes equal to the multiple of the pulse repetition rate in the train [9]. It is worth mentioning that transient coherent effects in ruby crystal at room temperature were studied in 1974 [10]. In that work, precessing magnetization was induced by a train of ultrashort pulses. Significant enhancement in magnetization was observed as the splitting between Zeeman sublevels of a Cr^{3+} ion became the multiple of the pulse repetition rate. However, no effect of Zeeman coherence on the absorptive properties of the crystal was studied.

To understand how the above theoretical description applies to ruby, let us consider the energy levels of Cr^{3+} in that crystal (see Fig. 2). Its ground state is described by the following spin Hamiltonian:

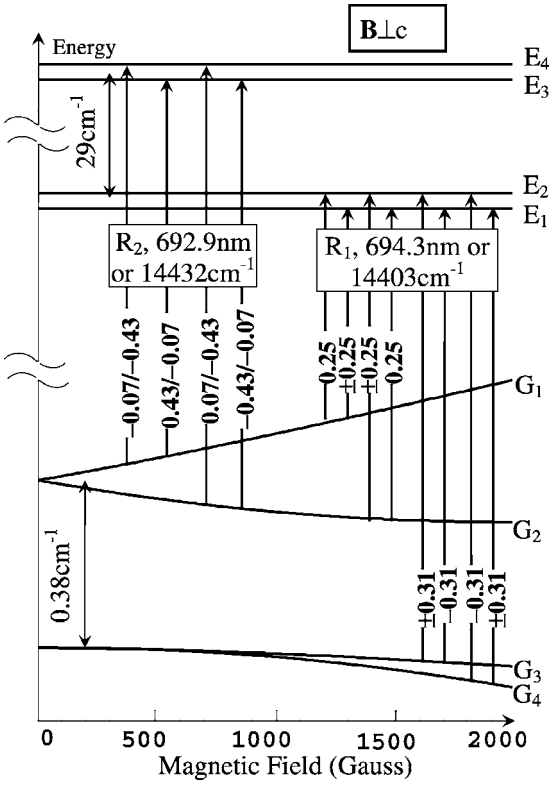

FIG. 2. Energy levels of Cr^{3+}-ion. Optical transitions allowed for σ-polarization of light propagating in a c direction with $\mathbf{B}\perp c$ are indicated by arrows connecting the corresponding energy levels accompanied by the corresponding matrix element. For R_1-line the upper (lower) sign stands for $\sigma^+(\sigma^-)$-polarizations respectively while for R_2-line the first and second number corresponds to $\sigma^+(\sigma^-)$ [11].

$$H = g_{\parallel}B_{\parallel}S_z + g_{\perp}\mathbf{B}_{\perp}\cdot\mathbf{S}_{\perp} + D[S_z^2 - S(S+1)/3], \qquad (2)$$

where $g_{\parallel}=1.982$ and $g_{\perp}=1.987$ are the ground-state g factors in the directions parallel and perpendicular to the crystal c axis, respectively, $B_{\parallel}$ and $\mathbf{B}_{\perp}$ are the corresponding magnetic field components, $2D=-11.47$ GHz (-0.38 cm^{-1}) is the zero-field splitting of the ground state, and $S=3/2$ is the spin of Cr^{3+}. The width of both optical transitions is 11 cm^{-1} while the oscillator strength is 7.5×10^{-7}.

The situation when $\mathbf{B}$ is perpendicular to the c axis is of particular interest. The matrix elements on Fig. 2 are normalized so that the sum of their squares for each of the $\sigma^{\pm}$ polarizations is unity. There are three pairs of Λ systems: the one connecting G_1 and G_2 through E_1 and E_2, another one connecting G_1 and G_2 through E_3 and E_4, and a third one connecting G_3 and G_4 through E_1 and E_2. For circularly polarized light the products of the optical matrix elements of both Λ systems in each pair have the same sign, i.e., both Λ systems tend to excite ground-state coherence in phases. This is not the case for linearly polarized light when the two Λ systems in each pair tend to excite Zeeman coherence with exactly the opposite phase. Thus, CPT can be expected only under illumination by circularly polarized light.

The experiments were performed on a $3\times 3\times 5$ mm^3 dilute ruby crystal (0.002% of Cr^{3+}) with the c axis being

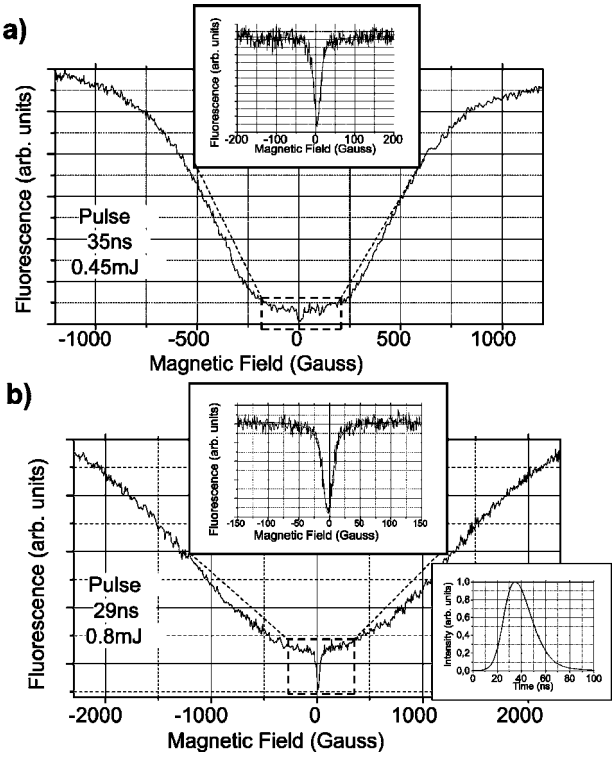

FIG. 3. CPT resonances in a zero magnetic field: (a) trace for the R_1 line and (b) trace for the R_2 line. Narrow peaks are shown in detail in the insets. See the text for details.

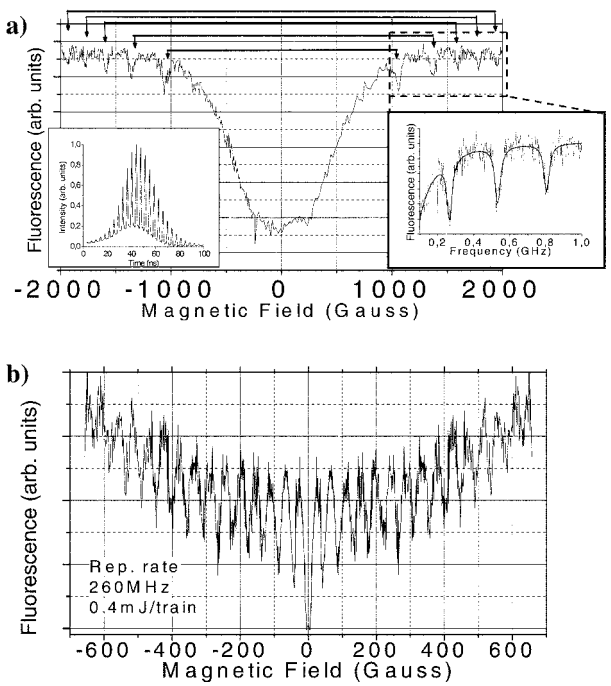

FIG. 4. CPT resonances with a mode-locked laser: (a) trace for the R_1 line and (b) trace for the R_2 line. See the text for details.

perpendicular to one of 3×5 mm^2 sides (Scientific Materials Corp.) A homemade tunable Ti:sapphire laser pumped by the doubled Nd YLF pulsed laser could operate both in a long pulse regime [pulse duration ≈30 ns full width at half maximum (FWHM)] and in a mode-locked regime delivering ≈30 ns pulse trains with repetition rate ≈260 MHz. The typical output energy per pulse or per train was several tenths of a *mJ* at a 100 Hz repetition rate. The laser polarization was linear. The typical shapes of pulses in both regimes are shown in the insets of Figs. 3 and 4. The shape of the pulse train was taken with a 350 ps rise-time photodiode and a 500 MHz bandwidth oscilloscope. The duration of each individual pulse in the train was not measured. The crystal was placed into a varying magnetic field with a $\mathbf{B}\perp c$ axis and illuminated along its axis by laser pulses sent through a $\lambda/4$ plate and focused by an $f=10$ cm lens. The fluorescence, i.e., the excited state population, was detected as a function of a slowly varying periodic magnetic field.

The experimental results for the case of long pulse excitation for both R_1 and R_2 lines are shown in Fig. 3. The spectra were averaged over several hundreds of magnetic field sweeps. In the case of the R_1 line, there are two features present in the spectrum: the broad one with the plateau on the bottom and a sharp one of a much smaller amplitude. The smaller one corresponds to less than a 0.5% decrease in the fluorescence amplitude and has a width of 18 G. It corresponds to a CPT resonance due to a $G_1\leftrightarrow E_1$, $E_2\leftrightarrow G_2$ Λ system, thus its width in terms of frequency units is ≈100 MHz since the g factor for the $G_1\leftrightarrow G_2$ transition is ≈4. The broad drop in the fluorescence signal has an amplitude ≈7–8%. Its FWHM is close to ≈950 G. The presence of a plateau at the bottom of that feature indicates that this is a CPT resonance in the $G_3\leftrightarrow E_1$, $E_2\leftrightarrow G_4$ Λ system with levels G_3 and G_4 being split very slowly as the magnetic field increases. Its width recalculated into the frequency domain is ≈40–50 MHz, i.e., this CPT resonance is, in fact, much sharper than the one discussed above. The possible explanation for this fact is as follows. The main contribution to the EPR linewidth of Cr^{3+} in ruby is known to originate from a magnetic dipole-dipole hyperfine interaction with neighboring ^{27}Al nuclei [12]. Thus, the magnetic-allowed Zeeman transition $G_1\leftrightarrow G_2$ should be broadened much stronger than the magnetic-forbidden $G_3\leftrightarrow G_4$. This also explains more than an order of magnitude difference in the amplitudes of those two CPT resonances, since in the Λ system with a longer-lived Zeeman coherence CPT should be more pronounced. It is worth mentioning that the spectroscopic information about the widths of magnetic-forbidden Zeeman transitions cannot be obtained in EPR measurements. Thus, the present results contain spectroscopic information about ruby.

For the R_2 line only one CPT resonance, represented by a sharp feature at the bottom of a smooth background, is observed. It has the width of 18 G, i.e., the same as the one observed at the R_1 line, thus it corresponds to the $G_1\leftrightarrow E_3$, $E_4\leftrightarrow G_2$ Λ system. The decrease in the fluorescence is 3–4% at the center of the resonance. The fact that this resonance is much stronger is easily understandable, since (1) there is no background fluorescence excited from the other pair of levels, G_3 and G_4, and (2) the transitions G_1, $G_2\leftrightarrow E_3$, E_4 are three times stronger than G_1, $G_2\leftrightarrow E_1$, E_2. The smooth background originates, probably, from the level mixing in high magnetic fields and consequent change in the optical selection rules. That was confirmed by removing the $\lambda/4$ plate,

i.e., by making laser light linearly polarized. Under this condition all three CPT resonances, discussed above, disappeared, as they were supposed to, while the smooth feature in question remained.

The results for trains of ultrashort pulses are shown in Fig. 4. CPT resonances corresponding to the $G_3 \leftrightarrow E_1$, $E_2 \leftrightarrow G_4$ Λ system of the R_1 line are not equidistant because of the nonlinear dependence of the levels G_3 and G_4 separation on the magnetic field strength. However, if recalculated into frequency domain, their positions exactly correspond to the harmonics of the laser repetition rate (260 MHz). The first three of them are shown in the inset of Fig. 4(a). Their widths (FWHM) are in the range 35–38 MHz in rather good agreement with the result obtained for the $G_3 \leftrightarrow G_4$ transition in a long pulse regime. It was not possible to observe multiple CPT resonances corresponding to the $G_1 \leftrightarrow E_1$, $E_2 \leftrightarrow G_2$ Λ system with an appreciable signal-to-noise ratio due to the very small amplitude of the CPT effect ($<0.5\%$) and the rather unstable operation of the laser in a mode-locked regime.

For the R_2 line, almost 30 CPT resonances are indicated [see Fig. 4(b)]. The magnetic field separation between each pair of resonances is $\approx$45 G. This value corresponds to 252 MHz of frequency separation which is somewhat close to 260 MHz of the laser repetition rate. Due to a rather poor signal-to-noise ratio and a rather low spectral resolution it is not possible to exactly determine the linewidth of resonances, but it seems to correspond rather well to the value of 18 G obtained in a long pulse regime.

In conclusion, a mechanism of CPT in a pulsed regime is described theoretically on the basis of a simple model. It relies on the slower optical excitation rate in the presence of coherence between ground-state sublevels rather than on populating of the dark state by population relaxation from the excited optical state as in conventional CPT experiments in a continuous wave (CW) regime. The experimental test of the proposed mechanism resulted in the observation of CPT in a crystalline solid (ruby) at room temperature. Zeeman coherence was excited in several percentages of all ions, a few orders of magnitude more than in the low-temperature experiments mentioned above. Spectroscopic information about the widths of magnetic-forbidden Zeeman transitions of Cr^{3+} in ruby, not accessible by other methods, is obtained. Possible applications of the reported phenomena include a room-temperature all-optical analog of EPR spectroscopy in low magnetic fields, suppression of excited state absorption in laser crystals [13], alternative technique of mode-locking in lasers [14], etc.

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