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Ultraslow light propagation in an inhomogeneously broadened rare-earth ion-doped crystal

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Abstract

We show that Coherent Population Oscillations effect allows to burn a narrow spectral hole (26 Hz) within the homogeneous absorption line of the optical transition of an Erbium ion-doped crystal. The large dispersion of the index of refraction associated with this hole permits to achieve a group velocity as low as 2.7 m/s with a transmission of 40 %. We especially benefit from the inhomogeneous absorption broadening of the ions to tune both the transmission coefficient, from 40 % to 90 %, and the light group velocity from 2.7 m/s to 100 m/s.

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Reduction of the optical group velocity by several orders of magnitude relatively to the speed of light propagating in vacuum was demonstrated in recent years in diverse media among which Bose-Einstein condensate, cold atoms [1, 2], vapors [3, 4] and solids [5, 6, 7, 8]. In addition to the strangeness of producing light propagating at speeds as low as few m/s, Slow Light Propagation (SLP) is at the very heart of new fundamental and applied fields of research in nonlinear and quantum optics. From the nonlinear optical side, SLP allows to strongly enhance the light-matter interaction time. Moreover, this interaction time can be continuously tuned to produce optical buffers and variable delay lines for optical networks. From the quantum optical point of view, SLP should allow, under specific conditions, classical and quantum properties of an electromagnetic field to be mapped into an atomic system [10]. The fundamental physical idea at the origin of SLP is the creation of a very narrow spectral hole in the homogeneous absorption profile. As stipulated by Kramers-Krönig relations, this narrow spectral hole is accompanied by a strong dispersion of the index of refraction inducing a low group velocity and an increase of the transmission. These two aspects are crucial in the choice of the atomic system and the coherent interaction inducing SLP.

The first direct demonstration of SLP [1, 2, 3, 4, 5] was achieved via Electromagnetically Induced Transparency (EIT) [9]. It was originally implemented by applying a secondary control field to eliminate the linear absorption of a resonant probe field through an otherwise absorbing medium. The standard scheme for EIT is a three-level Λ system, where the probe field drives the system from one of the ground states and the control field from the second ground state.

The width of the spectral hole burned in the homogeneous absorption profile is proportional to the inverse of the dephasing time of the ground state. Another physical effect recently used to reduce the speed of light to as low as few tens of meters per second is Coherent Population Oscillations (CPO) [6, 7, 8]. In contrast to EIT, the CPO [11, 12] effect is easily achieved in a two-level system which is excited by a "strong" pump field E_p oscillating at ω_p, along with a weak probe field E_s oscillating at $\omega_s = \omega_p + 2\pi\delta$. In this configuration, the two-level system population oscillates at the pump-probe beat frequency $\delta = (\omega_s - \omega_p)/2\pi$. Such oscillations become significant when δ is less than $1/T_1$, where T_1 is the relaxation lifetime of the excited state. As a result a narrow spectral hole, whose width is proportional to $1/T_1$, is induced in the homogeneous absorption profile of the probe field. M. S. Bigelow *et al.* [6] used this effect in a 7.25-cm-long ruby crystal at room temperature to reduce the group velocity of light down to 57 m/s with a spectral hole width of 36 Hz and a rather low total transmission of about 0.1 %. More recently, P.C. Ku *et al.* [8] showed in GaAs/AlGaAs semiconductor quantum wells a high transmission of about 50 % for a group velocity of 9600 m/s. Though this value is low compared to the speed of light in vacuum, it is still limited by the rather short relaxation time $T_1 \sim 1$ ns.

In this letter, we report on SLP induced by the CPO effect in a crystal insulator doped with rare-earth ions. Thanks to the long-lived excited state of the ions which is at the origin of the very narrow spectral hole and of the steep dispersion of the refraction index, we successfully achieved ultraslow light propagation in a solid-state material. The corresponding group velocities are comparable to those achieved in SLP via EIT in a Bose-Einstein condensate [1, 2]. The optical intensities we use are as weak as the intensities concerned in Ref. [1, 2] and are orders of magnitude weaker than those used in Ref. [6, 8] for SLP via the CPO effect. In contrast to Ref. [6, 7, 8], we investigate in details the influence of the power and the absorption coefficient on the SLP. More importantly, we show that the inhomogeneous broadening inherent to rare-earth ion-doped crystals, constitutes an additional control parameter we are able to manipulate to tune both the group velocity and the transmission coefficient.

The crystal used in our experiments is supplied by Scientific Materials Inc., and it consists in an Yttrium oxyorthosilicate (Y_2SiO_5) lattice doped with 0.005 at. % of $^{167}Er^{3+}$ ions. The optical transition between the fundamental state $^4I_{15/2}$ and the excited state $^4I_{13/2}$ of the Erbium ions is at 1536.1 nm. The crystal temperature is maintained at 1.5 K in a liquid Helium cryostat in order to reduce the homogeneous linewidth to the sub-MHz level. However, due to the static electric field strength in the crystal an inhomogeneous broadening of about $\Gamma_{\text{inh}} = 1.3$ GHz is present and prevents from the direct measurement of the homogeneous spectral line. Using the spectral hole burning method, we measure the lifetime T_1 of the excited state to be 8 ms. Photon echo technique is used to measure the dephasing time T_2 which is found to be about 3 μs at

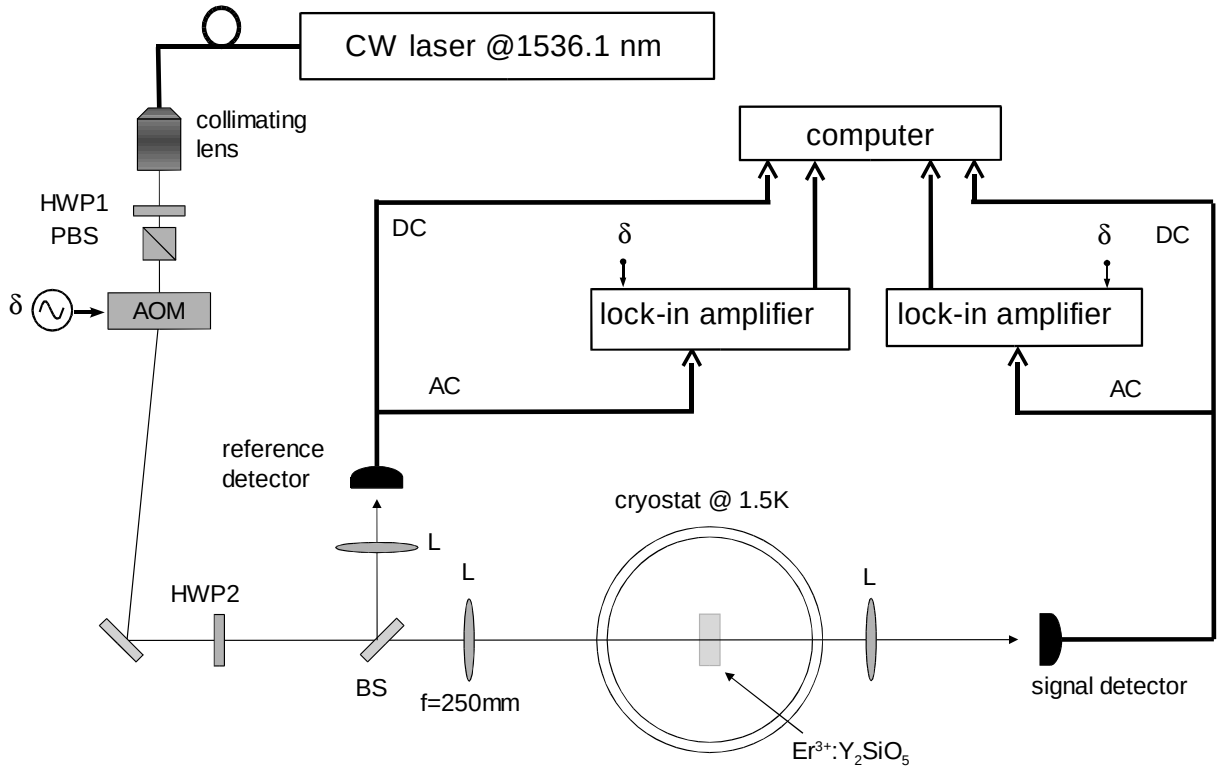


Figure 1: Scheme of the experimental setup: HWP1, HWP2 half-wavelength plates; PBS polarizing beam splitter; AOM acousto-optical modulator driven by a sinusoidal wave function oscillating at δ; BS beam splitter; L lens; AC Alternative Current; DC Direct Current.

1.5 K, a rather short value in comparison with T_1.

From the theoretical point of view, the pump beam allows to selectively excite a narrow portion of ions within the inhomogeneous line: those in resonance with the pump frequency. Such ensemble of ions can be described by the formalism developed in Ref. [6, 11], where a two-level system is driven by $E_p e^{-\imath\omega_p t} + E_s e^{-\imath\omega_s t}$ with $\omega_s = \omega_p \pm 2\pi\delta$. Following the mathematical analysis in Ref. [6, 11], expressions of the absorption $\alpha(\delta)$ and the index of refraction $n(\delta)$ are derived for $T_2 << T_1$ from the off diagonal density matrix elements oscillating at δ,

$$\alpha(\delta) = \frac{\alpha_\circ}{1+S}[1 - \frac{S(1+S)}{(2\pi\delta T_1)^2 + (1+S)^2}], \tag{1}$$

and

$$n(\delta) = 1 + \frac{\alpha_\circ c T_1}{2\omega_p}\frac{S}{1+S}\frac{2\pi\delta}{(2\pi\delta T_1)^2 + (1+S)^2}. \tag{2}$$

The different parameters involved in Eqs. (1) and (2) are the saturation parameter S defined as the ratio of the pump intensity and the saturation intensity and the unsaturated absorption coefficient $\alpha_\circ$ of the two-level system. The Half Width at Half Maximum (HWHM) of the spectral hole induced by the CPO effect is theoretically given by

$$\Gamma_{\mathrm{HWHM}} = \frac{1}{2\pi T_1}(1+S). \tag{3}$$

The main concern of this letter is the determination of the group velocity v_g experienced

by the probe field propagating in the crystal of length L. It is deduced from the time delay τ accumulated by the probe, relatively to a reference beam. The group velocity is given by $v_g = L/\tau$, where a theoretical expression of the delay is easily obtained from Eq. (2):

$$\begin{aligned}\tau(\delta) &\simeq \frac{L}{c}\frac{\omega_p}{2\pi}\frac{n(\delta)-n(-\delta)}{2\delta} \\ &= \frac{\alpha_\circ T_1 L}{2}\frac{S}{1+S}\frac{1}{(2\pi\delta T_1)^2+(1+S)^2},\end{aligned} \tag{4}$$

where we use the fact that $n \simeq n_0 + (\omega_p/2\pi)\partial n/\partial\delta$.

According to Eq. (3), the width of the spectral hole associated with $T_1 = 8\,\mathrm{ms}$ is expected to be of the order of few Hz. In this case, the frequency of the probe field is so close to the pump frequency that the observation of the narrow spectral hole is prevented by the relative frequency fluctuations of the pump and the probe laser sources. However, it is possible to overcome this problem by generating both the pump and the probe fields from a single laser source whose intensity is modulated at a frequency δ with a $1 + m\cos(2\pi\delta t)$ wave function, where $m < 1$ is the modulation depth. Indeed, the strong mean value of the modulated laser signal plays the role of the pump whereas the Fourier sidebands of the modulation at $\pm\delta$ act as the weak probe. Such an approach has been successfully used in Ref. [6, 7].

The scheme of our experimental setup is depicted in Fig. 1. The light source we use is a 2-kHz linewidth, continuous-wave fiber laser emitting at 1536.1 nm. The combination of the half-wave plate HWP1 and the polarizing beamsplitter PBS allows to control the laser beam power. By driving the rf power of an acousto-optic modulator (AOM) with a $1+m\cos(2\pi\delta t)$ wave function, a 10 % intensity modulation depth is applied to the first order diffracted field. After the AOM, the beamsplitter BS allows to separate a reference beam from the main beam which is sent through the crystal located in the cryostat. Thanks to a 250-mm lens in front of the cryostat, the main beam is focused down to a waist of $540\,\mu$m in the 3-mm long crystal. The main and the reference beams are then detected using InGaAs photodiodes. The AC components of the photocurrents, proportional to the amplitude of the modulation, are demodulated using lock-in amplifiers. They are then stored in a computer, simultaneously with the DC components, after data sampling and data averaging over more than 10000 samples.

Figure 2 shows the measured delay τ of the weak sidebands for different modulation frequencies δ as a function of the pump power. The pump field wavelength is tuned to the maximum of the absorption in the inhomogeneous profile. The continuous lines are the plots of the theoretical expression Eq. (4) of the delay, where T_1=8 ms, L=3 mm. The unsaturated absorption coefficient at the maximum of the inhomogeneous line, $\alpha_\circ = 6.5\,\mathrm{cm}^{-1}$, is determined by measuring the linear absorption in the crystal of a weak laser beam as a function of its wavelength. Care is taken in tuning the frequency of the laser over few GHz ($> \Gamma_{\mathrm{inh}}$) within a sufficiently short time

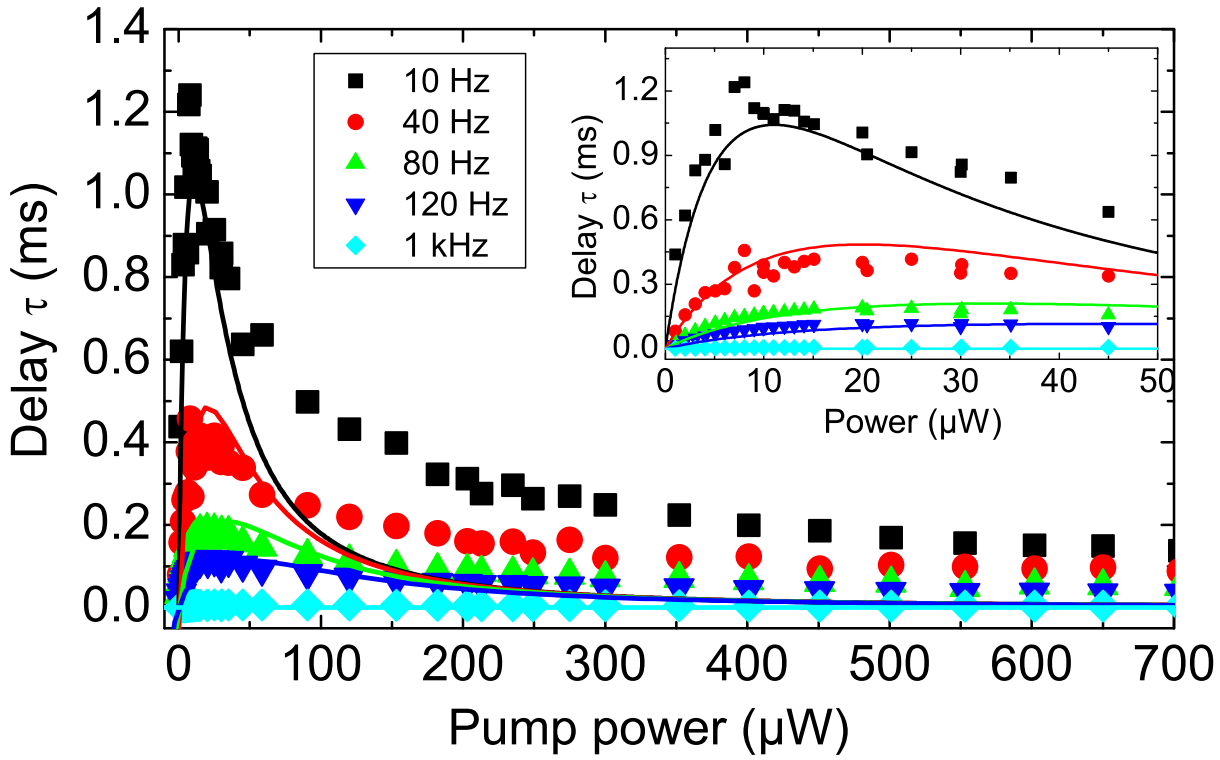


Figure 2: Experimental (points) and theoretical (continuous lines) delay τ of the probe as a function of the pump power for different modulation frequencies δ. Inset: zoom at low pump powers.

scale (5 ms) in order to avoid any saturation effect. As shown in Fig. 2, the delay τ tends to zero in the low pump power limit, $S << 1$, because the number of the excited ions is still not enough to induce the coherent effect. When the pump power is increased, $S >> 1$, the delay τ decreases as well because of the absorption saturation: the number of the excited ions contributing to the CPO effect is reduced and the spectral hole is power-broadened leading to a less sharp dispersion. The discrepancy between theory and experiment when $S >> 1$ [1] is probably due to the inhomogeneous broadening [12]. Indeed, it modifies the saturation behavior of the Erbium ions with respect to that of the homogeneously broadened two-level system considered in the model. A maximum delay τ_{max} is reached for an optimum value of $S(\delta)$ independent on $\alpha_\circ$. It is $S \simeq 1/2$ in the low frequency limit ($\delta < 1/T_1$) according to theory. Experimentally, $\tau_{max}^{exp} = 1.1 \pm 0.1$ ms at $\delta = 10$ Hz which is in a good agreement with the theoretical value $\tau_{max}^{the} = 1.04$ ms derived from Eq. (4). The delay τ_{max}^{exp} is obtained for a pump power of 12 μW measured in front of the cryostat which corresponds to an intensity of 2.1 mW/cm^2 on the crystal. In the following, we will work at this particular pump intensity in order to maximize the CPO effect. This intensity is rather low in comparison with former CPO experiments [6, 7, 8] where $\sim$kW/cm^2 are required, and is comparable to the intensities involved in SLP in Bose-Einstein condensate [1, 2]. The

[1] We experimentally checked that the increase of the sidebands intensity as the pump power is increased, is not responsible for the discrepancy in Fig. 2.

group velocity $v_g = L/\tau =$ associated with τ_{max}^{exp} is 2.7±0.2 m/s. To our knowledge, it is the slowest group velocity ever achieved in a solid. Indeed, according to our results, we reduce the group velocity by a factor $n_g = c/v_g = 10^8$. In comparison, the reduction is less than 5×10^6 in the Ruby and Alexandrite crystals [6, 7] and about 10^4 in the semiconductor quantum wells [8].

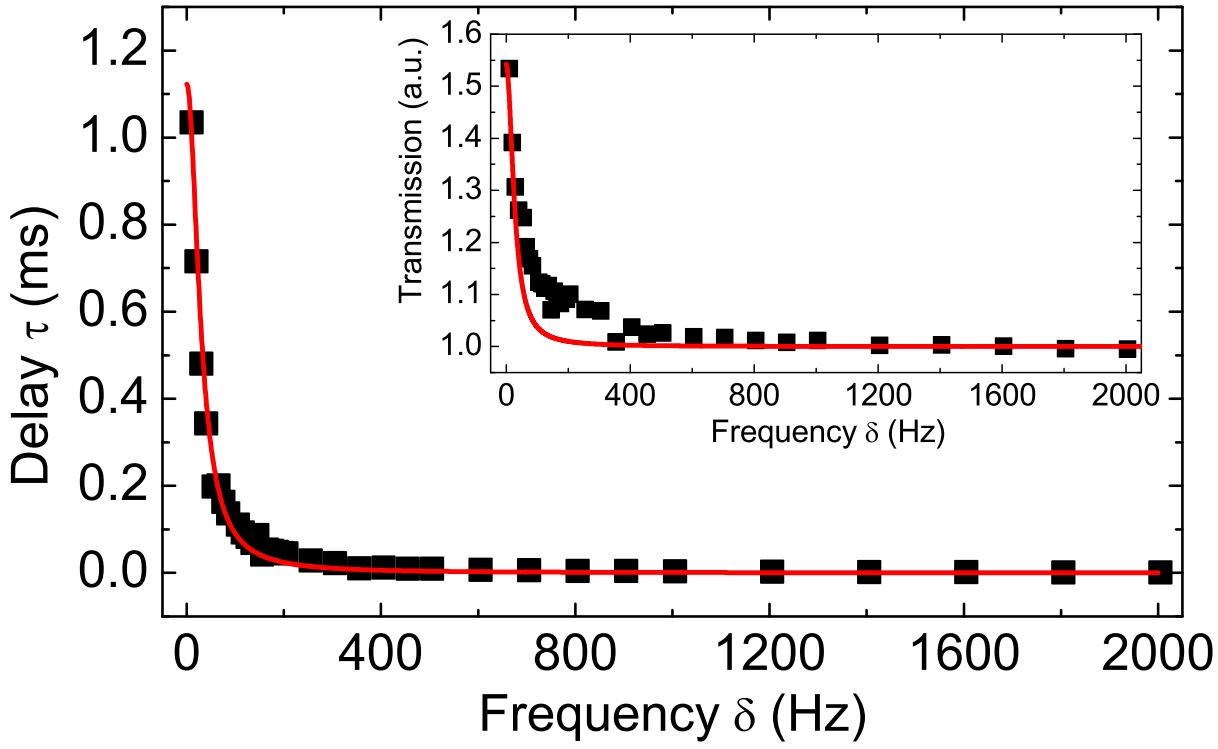


Figure 3: Experimental (squares) and theoretical (continuous lines) delay τ of the probe as a function of the modulation frequency δ and for a fixed pump power (12 μW) with S=1/2. Inset: the corresponding normalized transmission.

In order to determine the linewidth of the narrow spectral hole which is at the origin of the large dispersion of the refraction index and hence of the SLP, we measure the delay as well as the transmission of the weak sidebands as a function of the modulation frequency (the squares in Fig. 3). Like in the previous measurement, the wavelength of the pump is tuned to the maximum of the absorption. The inset in Fig. 3 is the transmission of the weak sidebands normalized to the transmission that is observed in a hole burning experiment. Indeed, a spectral hole is burned by the pump field in the inhomogeneous transmission profile of the probe field. Due to CPO effect, an additional increase of the transmission is observed when δ is smaller than $1/T_1$. This increase is about 55 % at δ= 10 Hz. At frequencies δ larger than $1/T_1$, the effect of CPO becomes negligible. The continuous lines in Fig. 3 are the theoretical plots derived from Eq. (1) and Eq. (4) without adjustable parameters. We can deduce from the inset in Fig. 3 a 26-Hz HWHM of the spectral hole, which is rather close to the theoretical value of 30 Hz.

One of the fundamental point we are addressing in this letter is the experimental exploration and exploitation of the dependance of the group velocity reduction on the absorption coefficient.

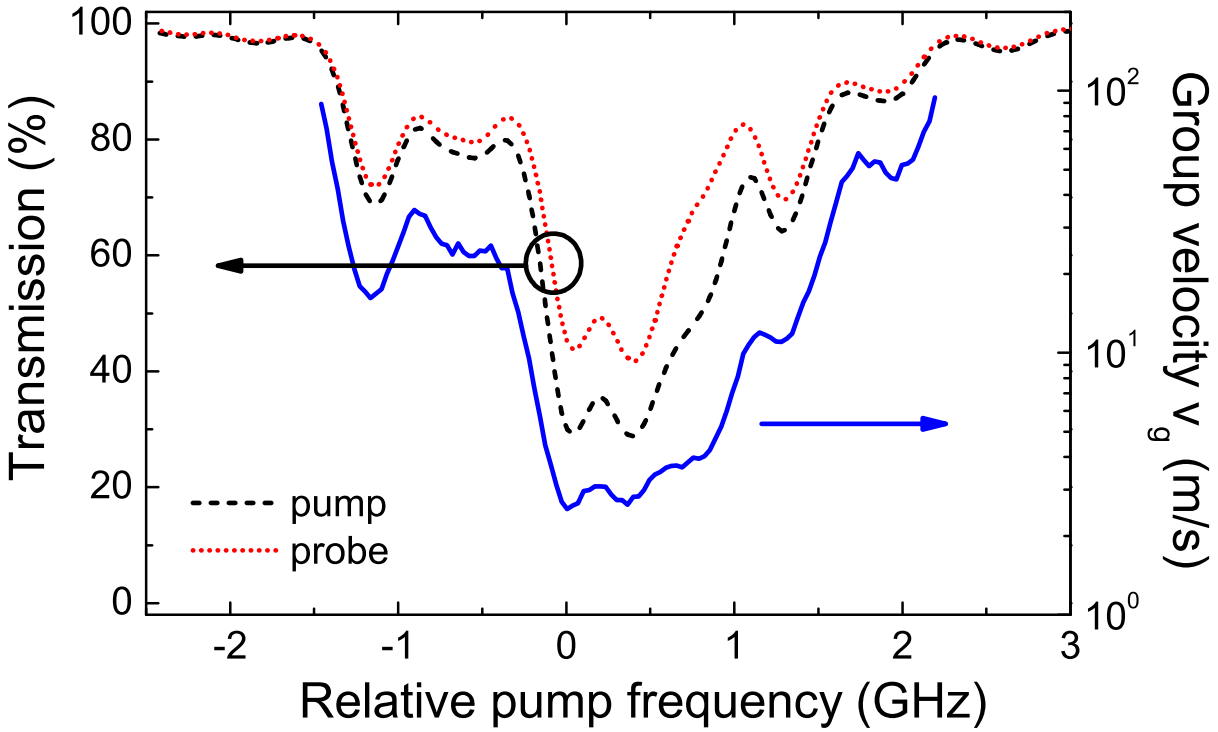


Figure 4: The experimental group velocity (continuous line), pump (dashed line) and probe (dotted line) transmissions as a function of the pump frequency. The modulation frequency δ is fixed at 10 Hz and the pump power is such that $S = 1/2$.

Indeed, unlike the other materials that have been investigated up to now [6, 7, 8], our crystal shows an inhomogeneous absorption broadening. By tuning the pump frequency over this inhomogeneous profile, we are able to address different Erbium ions concentrations and hence different absorption coefficients. In Fig. 4, the continuous line represents the measured light group velocity as a function of the pump frequency detuning relative to the pump wavelength at the maximum of the absorption. We also plot the transmission of the pump field (dashed line) as well as the transmission of the probe field (dotted line). The frequency of the probe is detuned by $\delta = 10$ Hz from the pump frequency. The shape followed by the group velocity is similar to that of the transmission, clearly indicating the dependance of v_g on the absorption strength in the Erbium-doped crystal. However, a careful analysis of the results shows that v_g is not proportional to the inverse of the absorption. This might be due to the inhomogeneous broadening. From the practical point of view, Fig. 4 indicates that the inhomogeneous broadening is an additional parameter which allows the engineering of the group velocity. Indeed, thanks to the inhomogeneous broadening in our crystal, v_g can easily be varied from 3 m/s to 100 m/s by tuning the pump wavelength. The highest SLP is achieved at the maximum of the absorption with a probe transmission of 40 %. Moreover, an important reduction of the group velocity is still obtained for high probe transmission. For example, when the relative pump frequency is set at 2 GHz (see Fig. 4), a group velocity of 58 m/s is measured with a transmission of about

88 %.

It is worth noting that the delay τ is proportional to the product $\alpha_\circ T_1 L$. Consequently, the reduction of the group velocity n_g varies like the product $\alpha_\circ T_1 c$. The slowest light propagation will be then obtained in long-lived atomic systems with high absorption coefficient. However, in order to obtain a non negligible transmission, $T \sim e^{-\alpha_\circ L}$, one has to compensate for the large coefficient $\alpha_\circ$ by choosing a short medium. It is for example possible to obtain a 88 % transmission in the Erbium-doped crystal with the lowest group velocity, 3 m/s, by using a crystal having a thickness of about 200 μm pumped at the maximum of the absorption profile [2]. This means that transparency can be reached while keeping the group velocity at its minimum value.

In conclusion, the implementation of CPO in Erbium-doped crystals enables to achieve ultra SLP. The pump intensities involved are six order of magnitude smaller than in former CPO experiments [6, 7, 8] and comparable to the one involved in EIT in ultracold atomic gas [1, 2]. We demonstrate that the inhomogeneous broadening, an inherent effect of the crystalline matrix, does not degrade the slow light propagation effect. It can instead be used to spectrally tune both the slow light propagation effect and the transmission. Group velocities ranging from 3 m/s to 100 m/s are achieved with transmission efficiencies ranging from 40% to 90%.

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