Saturable Absorber, Coherent Population Oscillations, 
and ”Slow Light”

V.S.Zapasskii and G.G.Kozlov

e-mail: gkozlov@photonics.phys.spbu.ru

All-Russia Research Center ”Vavilov State Optical Institute”, St. Petersburg, 199034 Russia

The paper presents a critical analysis of publications devoted to one of the methods for production of the so-called ”slow light” (the light with an anomalously low group velocity) due to extremely high local steepness of the dispersion curve of the medium. The method in point employs for this purpose the effect of coherent population oscillations accompanied by burning of a narrow spectral hole in the homogeneously broadened absorption spectrum. Physical model of the effect proposed in the studies under consideration is based on analysis of response of a nonlinear medium to a low-frequency intensity modulation of the incident light beam. We show that all the observations described in these papers can be easily interpreted in the framework of the simplest model of saturable absorber and have nothing to do with the effects of hole burning and group velocity reduction.
I. INTRODUCTION

After publication of the first impressive results on group velocity reduction by several orders of magnitude [1,2], the effects of slowing down, stoppage, storage, and release of light attracted attention of a great number of researchers. The experiment [2], which demonstrated the light velocity reduction by more than 7 orders of magnitude, was performed with sodium atoms at the temperatures close to that of the Bose-Einstein condensation (hundreds of nK) and was based on the effect of electromagnetically induced transparency. The paper [2] initiated a great number of publications, with some of them, in particular, claiming observation of the light velocity reduction of similar scale under much less exotic experimental conditions (e.g., at room temperature). To create a steep dispersion of the refractive index, along with the effect of electromagnetically induced transparency, there has been used the effect of coherent population oscillations (CPO). Observation of the CPO-based hole burning in the absorption spectra of ruby and alexandrite crystals was reported in [3,4]. Then, in [5,6], there has been announced observation, in these crystals, of ”slow” and ”fast” light, i.e., the light with ”ultralow” and, in this particular case, ”negative” group velocity. More recently, other publications have appeared that supported the proposed interpretation [7,8], and, as a result, the CPO effect was included into the arsenal of the ”slow light” methods (see, e.g., [9,10]).

The goal of this publication is to show that the light-pulse delay detected in the above experiments is a well known effect of nonlinear optics. It has nothing in common with the CPO effect and is not connected with the group velocity reduction in the medium. In our opinion, the experimental data of [3–6] provide no evidence for the ”slow light” observation, and their interpretation is erroneous.

II. THE EFFECT OF COHERENT POPULATION OSCILLATIONS

The role of population oscillations in the coupling of the light waves with close frequencies propagating in a resonant nonlinear medium was first noted, as far as we know, by W. Lamb in 1964 [11]. Analysis of the properties of a saturable absorber performed by Schwartz and Tan in 1967 [12] has shown that, in the presence of a strong (saturating) pump wave, a weak probe wave scanned in the vicinity of the strong wave frequency displays an apparent dip in the absorption spectrum with the center at the pump frequency and with the width corresponding to the inverse absorption relaxation time. The effect had a simple and visual physical interpretation. The combined action of two monochromatic waves, due to their interference, gives rise to a modulation of the total field intensity at the beat frequency. When this frequency appears to be sufficiently low (comparable with the inverse population relaxation time or lower), the light intensity modulation is transformed into modulation of
populations (and absorption) of the optically active centers. Under these conditions, interaction of the monochromatic pump wave with the oscillating susceptibility of the medium destroys its monochromaticity and gives birth to sidebands, with one of them being exactly coincident in frequency with the probe wave and thus contributing to its intensity. This is revealed as an apparent bleaching of the medium (or as a dip in the absorption spectrum) in the vicinity of the pump frequency. The width of such a dip is, as a rule, smaller than homogeneous width of the optical transition. For this reason, this effect was often considered as "hole burning" in a homogeneously broadened spectrum.

The interest to the CPO effect in 70s of the last century was stimulated, on the one hand, by its role in the laser emission dynamics and, on the other, by the possibility to use it for getting information about lifetimes of excited states in nonluminescent or rapidly relaxing systems. In those years, the CPO effect has been thoroughly studied both theoretically (see, e.g., [13,14]) and experimentally [15,16]. The experimental studies mainly demonstrated applicability of this spectroscopic phenomenon to kinetic measurements. In spite of the fact that this effect is linear with respect to the probe wave and is phenomenologically close to the well-known effect of hole-burning in the inhomogeneously broadened spectra, it results from the multiwave interaction and is, evidently, of essentially different nature.

The conclusion about observation of a narrow spectral dip ($\Delta \nu \approx 37$ Hz) in the homogeneously broadened absorption spectrum of ruby crystal was made in [3] based on analysis of the crystal's response to the low-frequency intensity modulation of the light beam (Ar$^+$-laser). The dip was treated as a usual spectral feature in the complex refractive index of the medium. In [5], to this narrow spectral dip, in conformity with the Kramers-Kronig relations, was assigned an extremely steep slope of the dispersion curve, and the observation of "slow light" with a group velocity of $57.5 \pm 0.5$ m/s was reported. Similar experiments performed with alexandrite crystals were reported in [4,6]. This crystal, behaved, at certain wavelengths, as an inverse saturable absorber (with the absorption growing with light intensity), and the CPO effect, in this crystal, resulted in "anti-hole burning" in the absorption spectrum. The experimental observations of [6] have been interpreted in terms of "negative" group velocity of light ("fast" light). These papers provided the basis for a new approach to the problem of implementation of the "slow" and "fast" light. To evaluate the novelty and validity of this interpretation, let us turn to basic properties of a saturable absorber.

III. SATURABLE ABSORBER

Characteristic properties of a saturable absorber attracted attention of researchers in the early laser epoch, in 60s, when the phthalocyanine dyes were found to be efficient for laser Q-switching. In those years, their temporal and intensity-related characteristics have been studied in sufficient detail [17-20]. The simplest saturable absorber is known to be well
modeled by a two-level system with a high dephasing rate $T_2^{-1}(T_2^{-1} >> T_1^{-1}, \Omega$, where $T_1$ is the population relaxation time and $\Omega$ is the Rabi frequency). As a good example of the saturable absorber may also serve ruby crystal (Fig. 1a). The energy structure of chromium ion in ruby is not two-level, but a high relaxation rate of the excited state $^4F_2$ ($|b\rangle$ in Fig. 1) to the metastable levels $2\bar{A}$ and $\bar{E}$ ($|c\rangle$) makes it possible to neglect the coherent transient processes practically at any reasonable excitation power.

In the general case (neglecting the effects of propagation), dependence of intensity of the light transmitted through a saturable absorber $I_{out}$ on its intensity at the entrance $I$ can be presented in the form

$$I_{out} = \alpha(I, t)I,$$  

(1)

where the transmissivity factor $\alpha(I, t)$ controls all the dynamic and intensity-related characteristics of the saturable absorber.

Under nonstationary conditions, when the light intensity varies in time, the dynamics of transmission of the saturable absorber is described by the equation

$$\dot{\alpha} = \frac{\alpha_{eq} - \alpha}{\tau},$$  

(2)

Here, $\alpha_{eq}$ is the equilibrium value of $\alpha$ corresponding to the current light intensity, and $\tau$ is a time constant (at low light intensities $\tau \approx T_1$. When the light intensity is not too high, the dependence $\alpha_{eq}(I)$ can be presented in the form

$$\alpha_{eq}(I) = \alpha_0 - \alpha_1 I, \quad \alpha_1 I << \alpha_0.$$  

(3)

As the light intensity increases, the nonlinear is usually bleached. In this case, the parameter $\alpha_1$ is negative. There are situations, however, when absorption of the optical medium increases with increasing light intensity, rather than decreases. This situation is typical for certain schemes of optical orientation and is realized, in particular, in the alexandrite crystal mentioned above, where, due to efficient excited-state absorption, chromium ions in the excited metastable state may absorb light, at the wavelength of excitation, stronger than those in the ground state (Fig. 1b). For these *inverse* saturable absorber, $\alpha_1 > 0$.

Solving Eqs. (1)–(3) for a stepwise change of the incident light intensity, one can easily make sure that the time dependence of the transmitted light intensity, in this case, contains two components - an inertialless (stepwise) jump and an exponential component, reflecting dynamics of establishment of steady-state populations of the medium. The sign of the exponential component, evidently, depends on the sign of $\alpha_1$ (Fig. 2). In virtue of linearity of the Fourier-transform, the appropriate response in the frequency domain will also contain two components - a frequency-independent ("white") pedestal and a Lorentz-wise peak in the range of zero frequencies with a width of $\sim 1/\tau$. This dependence can be easily obtained...
in analytical form by calculating the response of the saturable absorber to the light beam with harmonically modulated intensity

\[ I = I_0 + I_1 \exp(i\omega t), \]  

(4)

From Eqs. (1)–(3), neglecting the terms \( \sim I_1^2 \), we have:

\[ I_{\text{out}}(t) = I_0(\omega_0 - \omega_1 I_0) + I_1 \exp(i\omega t)K(\omega) \]  

(5)

\[ K(\omega) \equiv \omega_0 - \omega_1 I_0\left(1 + \frac{1}{1 + i\omega \tau}\right) = |K(\omega)|e^{i\phi} \]  

(6)

Here, as usual, the complex intensities \( I \) and \( I_{\text{out}} \) describe the amplitude-phase relations between harmonically oscillating light intensities at the entrance and at the exit of the absorber.

Figure 3 shows characteristic frequency dependences of the amplitude \( K(\omega) \), phase \( \phi(\omega) \), and time delay \( \delta(\omega) = \phi(\omega)/\omega \) of intensity oscillations of the light beam transmitted through a usual (bleachable) saturable absorber (a) and through an inverse saturable absorber (b).

As is seen from these curves, the greatest time delay is experienced by low-frequency spectral components of the light intensity (\( \omega << \tau^{-1} \)), for which, taking into account the condition \( \omega_1 I_0 << \omega_0 \) from Eq. (3), we have

\[ \Delta t_{\text{max}} = \Delta t(\omega \rightarrow 0) = -\frac{\omega_1 I_0}{\omega_0} \]  

(7)

This formula yields the time delay experienced by a smooth pulse with the width much larger than the relaxation time \( \tau \). As expected, the time delay may constitute only a small fraction of the pulse width.

Figure 4 shows normalized light pulses at the entrance (solid lines) and at the exit (dashed lines) of the usual (a) and inverse (b) saturable absorber for several ratios of the pulse width \( \delta \) and relaxation time \( \tau \). As seen from the figure, the pulse propagating through a bleachable absorber generally experiences positive time delay, whereas in an inverse absorber the delay is negative. Note once again that all these results have been known for several decades (see, e.g., paper [18], in which the shape of the pulse transmitted through a saturable absorber is calculated). Let us now turn back to the experiments on observation of ”slow light” under conditions of coherent population oscillations [5,6].

IV. DISCUSSION

Comparison of the results presented above with the experimental data of [3–6] did not allow us to find any noticeable distinctions between them. Specifically, in full agreement
with the model of saturable absorber, the amplitude of the light intensity modulation at the
exit of the absorber shows a Lorentz-wise peak in the region of low frequencies \( \omega \lesssim 1/\tau \) (Fig. 3 from [3]), and the phase shift of the intensity modulation, at these frequencies, is positive
or negative depending on the type of the absorber (Fig. 3 from [5] and Fig. 2 from [6]). A
positive time delay is experienced also by Gaussian pulse transmitted through a bleachable
absorber (cf. Fig. 4 from [5] and Fig. 4 of the present paper).

Thus, all the observations of [3–6], including ”slow” and ”fast” light, can be interpreted
in a fairly trivial way, in the framework of the simplest model of saturable absorber. This
model, evidently, does not imply either burning of a narrow spectral hole or any modification
of the light group velocity in the medium.

The authors of [3–6] interpret their experimental results in a more complicated way by
representing the harmonically modulated light beam in the form of three spectral compo-
nents, with the central one playing the role of the pump and the sidebands playing the role
of the probe. Such an approach has been repeatedly used to describe interaction of a
modulated monochromatic light with a saturable absorber and is, in principle, quite correct
[14,21]. First of all, however, the authors of [5] analyze the response of the absorber only
to one of the two side components and do not take into account their inevitable influence
upon each other (see, e.g., [21]). As follows from the model of the CPO effect, when the
probe beam contains two components symmetric with respect to the pump frequency, each
of them contributes to the other one and, in turn, experiences the influence of the latter to
its own amplitude and phase. Without considering this fact, the suggested theoretical model
is, in our opinion, inadequate. It is also incorrect to apply the standard notion of group
velocity to an absorbing medium with a strong spectral dependence of absorption (under
these conditions, the spectrum and shape of the light pulse should exhibit strong changes,
and the notion of group velocity needs to be redefined).

However, the main drawback of papers [3–6] is that the ”dip” detected in the space of
difference frequencies (beat frequencies between ”pump” and ”probe”) is interpreted as
a dip in the optical spectrum, which was not really observed and, under conditions of the
described experiments, could not be observed. To detect the dip, spectral width of the pump
beam (as well as of the probe) should be smaller than that of the dip. As applied to the case
of ruby crystal with a 37-Hz width of the ”dip”, the frequency of laser light should be stable
to within 14 (!) decimal digits (at least for the times of the order of inverse width of the
dip). In the experiments [3–6], the widths of the laser sources evidently exceeded the widths
of the ”burned” features by many orders of magnitude. Under these conditions, position of
the above ”narrow dip”, having no preferences in a wide spectral range, becomes physically
uncertain and loses any sense.

All the aforesaid is also true for other studies in which the retarded dynamics of a
saturable absorber was attributed to the hole-burning effect under conditions of CPO [7,8].
A different experimental situation was realized in [22], where as the pump and the probe were used quasi-monochromatic beams of independent laser sources, and the dip in the homogeneously broadened spectrum of excitonic absorption of a semiconductor structure with quantum wells was really detected in a classical way as was implied in early theoretical papers on the CPO effect (see, e.g., [14]). Based on the results of the amplitude and phase measurements, the authors concluded that the group velocity in the system under study, due to a high steepness of the dispersion curve in the region of the above dip, should be 9,600 m/s. This conclusion is, in our opinion, not sufficiently well grounded. First of all, in view of the already mentioned mutual influence of symmetric (with respect to the pump frequency) spectral components of the light pulse, the character of its interaction with the medium should crucially depend on its phase structure. In particular, as has shown our preliminary analysis, for a certain phase profile of the probe pulse, it proves to be insensitive to the action of the pump and incapable of detecting the dip. This fact indicates again the nonlinear nature of the "hole" detected in the CPO effect and incorrectness of direct application of the Kramers-Kronig relations to this spectral feature. And, lastly, as was already mentioned, it is inconsistent to apply the notion of the group velocity to the absorbing medium with a strong spectral dependence of imaginary part of the refractive index.

V. CONCLUSION

The results of the above treatment allow us to conclude that the experimental data of the papers [3–6], in which the "slow" or "fast" light was allegedly observed under conditions of the coherent population oscillations, can be easily interpreted in the framework of the simplest model of saturable absorber. In our opinion, the light pulse delay detected in these experiments have nothing to do with the hole- (or anti-hole-) burning in a homogeneously broadened absorption spectrum, as well as with reduction of the group velocity of light in the medium. These delays are caused, in fact, by self-induced distortion of the pulse propagating in the medium with intensity-dependent absorption. As for the CPO-based hole-burning effect, it can be observed only under conditions of quasi-monochromatic pump with its spectral width smaller than the inverse population relaxation time. To observe modification of the light group velocity in the medium under condition of the coherent population oscillations, the probe pulse spectrum should have approximately the same width. To the present day, to the best of our knowledge, no experiment of this kind has been performed. One may assume that, upon narrowing of the pump and probe pulse spectra, the contribution to the pulse delay related to the refractive-index dispersion feature of the medium in the region of the pump frequency will become noticeable on the background of the effects of retarded absorption considered in this paper. However, taking into account that this effect can be observed only in the absorbing medium, and the time delay of the pulse
may constitute only a small fraction of its width [9], this "slow light" will not differ from the "slow light" obtained using a saturable absorber, and the prospects of its application for practical purposes seem highly doubtful.

The authors are grateful to E.B.Aleksandrov for useful discussions.

[1] Casapi A., Jain M., Yin G.Y., and Harris S.E. //Phys. Rev. Lett. 1995. V. 74. P. 2447.
[2] Hau L.V., Harris S.E., Dutton Z., and Behroozi C.H. //Nature. 1999. V. 397. P. 594.
[3] Hillman L.W., Boyd R.V., Krasinski J., and Stroud C.R., Jr. //Opt. Commun. 1983. V .45. P. 416.
[4] Malcuit M., Boyd R.W., Hillman L.W., Krasinski J., and Stroud C.R. Jr. //J.Opt.Soc.Am. B. 1984. V. 1. P. 73.
[5] Bigelow M.S., Lepeshkin N.N., and Boyd R.W. //Phys.Rev.Lett. 2003. V. 90. P. 113903.
[6] Bigelow M.S., Lepeshkin N.N., and Boyd R.W. //Science. 2003. V. 301. P. 200.
[7] Baldit E., Bencheikh K., Monnier P., Levenson A., and Rouget V. //LANL, arXiv: ccsd-00004377, 2005.
[8] Zhang Yun-Dong et al. //Chinese Phys. Lett. 2004. V. 21. P. 87.
[9] Boyd R.W., Gauthier D.J., Gaeta A.L., Willner A.E. //Phys. Rev. A. 2005. V. 71. P. 023801.
[10] Matsko A.B., Strekalov D.V., and Maleki L. //Optics Express. 2005. V. 13. P. 2210.
[11] Lamb W.E. Jr. //Phys.Rev. A. 1964. V. 134. P. 1429.
[12] Schwarz S.E. and Tan T.Y. //Appl.Phys.Lett. 1967. V. 10. P. 4.
[13] Baklanov E.V. and Chebotaev V.P. //Zh. Eksp. Teor. Fiz. 1971. V. 61. P. 922.
[14] Sargent M. //Physics Reports. 1978. V. 43 P. 223.
[15] Song J.J., Lee J.H., and Levenson M.D. //Phys.Rev. A. 1978. V. 17. P. 1439.
[16] Lee J.H., Song J.J., Scarparo M.A.F., and Levenson M.D. //Opt.Lett. 1980. V. 5 P. 196.
[17] Gires F. and Combaud F. //J.Phys. 1965. V. 26. P. 325.
[18] Selden A.C. //Brit.J.Appl.Phys. 1967. V. 18. P. 743.
[19] Selden A.C. //J.Phys. D : Appl.Phys. 1970. V. 3 P. 1935.
FIGURE CAPTIONS

Fig. 1. Simplified energy-level diagrams of Cr$^{3+}$ ion in the ruby (a) and alexandrite (b) crystals. Under conditions of sufficiently strong optical pumping in the $|a\rangle - |b\rangle$ channel, a fast relaxation of electronic excitation from state $|b\rangle$ to metastable state $|c\rangle$ redistributes populations of the $|a\rangle$ and $|c\rangle$ levels (within the time intervals of the order of $\tau$) and changes the absorption of the crystal at the wavelength of excitation.

Fig. 2. Time dependence of intensity of the light beam transmitted through the usual (b) and inverse (c) saturable absorber upon step-wise change of the incident light intensity (a).

Fig. 3. Typical frequency dependences of the amplitude $K(\omega)$, phase $\phi(\omega)$, and time delay $\Delta t(\omega) = \phi(\omega)/\omega$ of intensity oscillations of the light beam transmitted through the usual (a) and inverse (b) saturable absorber.

Fig. 4. Normalized light pulses at the entrance (solid lines) and at the exit (dashed lines) of the usual (a) and inverse (b) saturable absorber for several ratios of the pulse width $\delta$ and relaxation time $\tau$. Calculations were performed for the pulse propagating in the presence of a background illumination (under conditions close to these of [5]).
FIG. 1. Simplified energy-level diagrams of Cr$^{3+}$ ion in the ruby (a) and alexandrite (b) crystals. Under conditions of sufficiently strong optical pumping in the $|a\rangle - |b\rangle$ channel, a fast relaxation of electronic excitation from state $|b\rangle$ to metastable state $|c\rangle$ redistributes populations of the $|a\rangle$ and $|c\rangle$ levels (within the time intervals of the order of $\tau$) and changes the absorption of the crystal at the wavelength of excitation.

\[ \Omega_1 \]

$\Gamma_{bc} \sim T_2^{-1}$

$\Gamma_{ca} \sim \tau^{-1}$
FIG. 2. Time dependence of intensity of the light beam transmitted through the usual (b) and inverse (c) saturable absorber upon step-wise change of the incident light intensity (a).
Fig. 3. Typical frequency dependences of the amplitude $K(\omega)$, phase $\phi(\omega)$, and time delay $\Delta t(\omega) = \phi(\omega)/\omega$ of intensity oscillations of the light beam transmitted through the usual (a) and inverse (b) saturable absorber.
FIG. 4. Normalized light pulses at the entrance (solid lines) and at the exit (dashed lines) of the usual (a) and inverse (b) saturable absorber for several ratios of the pulse width $\delta$ and relaxation time $\tau$. Calculations were performed for the pulse propagating in the presence of a background illumination.