Incoherent "Slow and Fast Light"

Zapasskii V.S. and Kozlov G.G.
Institute of Physics, St.-Petersburg State University, St.-Petersburg, 198504 Russia.
e-mail: gkozlov@photonics.phys.spbu.ru

We show experimentally that the effects of "slow and fast light" that are considered to be caused by spectral hole-burning under conditions of coherent population oscillations (CPO) can be universally observed with incoherent light fields on objects with the pure-intensity nonlinearity, when such an interpretation is inapplicable. As a light source, we used an incandescent lamp and as objects for study, a photochromic glass and a thermochromic coating. The response of the objects to intensity modulation of the incident light reproduced in all details the commonly accepted experimental evidences of the "light with a negative group velocity" and "ultraslow light". We come to conclusion that so far there are no experimental works providing evidence for real observation of the "CPO-based slow or fast light".

I. INTRODUCTION

Among the immense multitude of the effects of nonlinear optics, there may be singled out a group of phenomena comprehensively described in terms of light intensity without using explicitly the notion of the light field and its characteristics (frequency, spectral width, etc.). In these cases, the relation between the output light intensity \( I_{\text{out}} \) and intensity of the incident light \( I_{\text{in}} \) may be presented in the general form as

\[
I_{\text{out}} = K(I_{\text{in}}, t)I_{\text{in}},
\]

(1)

A characteristic feature of these effects, which are not associated directly with nonlinear polarizability of the medium at optical frequencies, is their certain sluggishness controlled by the recovery time \( \tau \) of the parameter \( K \). In the linear approximation, the dynamics of this recovery upon variation of the light intensity can be described by the simple differential equation

\[
\frac{dK}{dt} = \frac{K_{\text{eq}} - K}{\tau},
\]

(2)

where \( K_{\text{eq}} \) is the equilibrium value of the coefficient \( K \) for the current intensity \( I_{\text{in}} \). For relatively small variations of the light intensity \( I_{\text{in}} \), the dependence \( K_{\text{eq}}(I_{\text{in}}) \) can be approx-
imated by the linear expansion

\[ K_{eq}(I_{in}) = K_0 + K_1 I_{in}, \quad K_1 I_{in} << K_0, \] (3)

where the positive and negative signs of the coefficient \( K_1 \) correspond, respectively, to the cases of super- and sublinear dependences \[1\].

Equations (1) - (3), in spite of their simplified form, allow one to describe all the main features of response of such a nonlinear medium to variations of the incident light intensity (Fig. 1) \[1\]. In particular, temporal response of \( I_{out} \) to stepwise change of \( I_{in} \) exhibits exponential relaxation to its equilibrium value (Fig. 1a); the frequency dependence of the response (\( I_{out} \)) to a weak intensity modulation of \( I_{in} \) shows a Lorentzian feature with a half-width \( \sim 1/\tau \) peaked at zero frequency (Fig. 1b); and the appropriate frequency dependence of the modulation phase of \( I_{out} \) exhibits maximum delay (positive or negative depending on the sign of \( K_1 \)) at the frequency \( \sim 1/\tau \) (Fig. 1c). A pulse of the light modulation, in the general case, appears to be distorted. However, at low modulation frequencies, the absolute value of the time delay becomes frequency-independent, and, as a result, a sufficiently long smooth pulse experiences a pure shift with no reshaping (Fig. 1d). The sign of this shift may be also either positive or negative depending on the sign of \( K_1 \) (for more detail see \[1\]). One can notice that solutions of Eqs. (1) - (3) (Fig. 1) demonstrate alterations in the intensity spectrum of the light resulting from interaction with the nonlinear medium. For a spectroscopist, the easiest way to make it sure is to make use of the light with a ”white” intensity spectrum, i.e., the light with its intensity modulated by ”white” noise). Then, at the exit of the medium, the intensity spectrum of the light, initially flat, will display a peak (or dip) with a maximum at zero frequency and with the width \( \sim 1/\tau \). Usually, however, the frequency dependences of this kind are obtained using the light with a ”monochromatic” intensity spectrum: the intensity \( I_{in} \) is subjected to a weak harmonic modulation, with the amplitude and phase of \( I_{out} \) being measured at the exit of the sample at different frequencies.

The optical media with the above type of nonlinearity are usually referred to as saturable absorbers. The superlinear and sublinear dependences \[1\] correspond to two types of saturable absorbers, namely to the bleachable and so-called inverse absorbers. In the simplest cases, the optical nonlinearity \[1\] is the result of light-induced changes in populations of eigenstates of the system, with the characteristic recovery time \( \tau \) being determined by the population relaxation rate. Sometimes, the nonlinearity of this kind arises in optical systems
capable of transforming nonlinear variations of geometrical or polarization characteristics of
the beam into variations of the intensity signal, e.g., upon formation of the Kerr-type lens
in the systems with vignetting elements or upon light-induced polarization plane rotation in
the systems with polarizing elements. Such systems are sometimes referred to as artificial
saturable absorbers. The intensity-type nonlinearities can be also observed in reflection, in
scattering, in atomic systems under conditions of optical orientation, in impurity crystals
and glasses, in bulk and low-dimensional semiconductors, upon photochemical and photo-
conformation processes, upon light-induced heating of the medium, and so on, and so forth.
Moreover, Eqs. (1) - (3) containing no carrier frequency and, generally, not implying its
presence have no optical specifics and, evidently, may describe a much wider class of non-
linear physical problems. In [2], in particular, this circumstance has been demonstrated by
an example of an electric circuit with a nonlinear resistor.

All the aforesaid actually refers to well-known effects of nonlinear optics (or, better to
say, of nonlinear physics). In particular, the effect of the light pulse delay in a saturable
absorber is known for more than 40 years [3], and the media with intensity nonlinearity are
widely used starting from 60s in passive systems of Q-switching and mode-locking [4, 5]. In
the end of the 20th century, however, the above effects of retarded response of a saturable
absorber have been rediscovered [6, 7] and, without mentioning the known mechanism of
their origination, were ascribed to the light-induced changes in the group velocity of light in
saturable absorbers. As is known, the group velocity of light pulse with the carrier frequency
$\omega_0$ in a dispersive medium

$$V_{gr} = \frac{c}{n + \omega_0 dn/d\omega}$$

(4)
depends on steepness of the refractive index spectral dependence $dn/d\omega$ and, in principle,
may strongly differ from the "phase" velocity of light $c/n$. In the proposed interpretation,
the great value of the dispersion term in Eq. (4) was considered to be a consequence of a
narrow dip in the spectrum of the saturable absorber burnt by the monochromatic pump
wave due to the effect of coherent population oscillations (CPO) [8]. In other words, the
narrow peak observed at low frequencies in the intensity spectrum of the light transmitted
by the nonlinear medium was now regarded as lying in the optical spectrum of the medium.
Evidently, the new interpretation of the pulse delay in a saturable absorber invoking the
effects of propagation substantially differed from the previous interpretation and, in the
general case, could not be equivalent to the latter. Thereafter, this new idea, in spite of
its being seriously criticized (see [1, 9, 10]), has gained a wide popularity. There have been published dozens of papers on observation of such a "slow" and "fast" light, and the CPO effect turned into one of the basic technologies of this field of optics (see, e.g., [11, 12] and references therein). Such an interpretation of the effects of intensity nonlinearity proved to be viable probably because all the known experiments in this field were performed using laser sources, thus providing a preferential optical frequency needed to position the narrow spectral dip in its vicinity. In addition, all the theoretical papers were also based on the model of monochromatic electromagnetic fields. In this paper, we show experimentally that the effects of the so-called "slow" and "fast" light are universally observed in the incoherent light on non-laser sources on arbitrary objects with the intensity-type nonlinearity. As the light source, we used an incandescent lamp, with the object for study being a photochromic glass and a decorative thermochromic coating. Both objects were characterized by rather long relaxation times (∼10 s), which also provided a relatively low threshold of the optical nonlinearity. The time delays of intensity modulation of the light after interaction with the medium demonstrated what is usually considered as the "light with negative group velocity" and "ultraslow light" though the CPO-based model of the group velocity modification, in this case, cannot be valid.

II. "NEGATIVE GROUP VELOCITY OF LIGHT" IN PHOTOCHROMIC SUN GLASSES

The first object for study was a photochromic spectacle glass (Opticoelektron group JSC - Panagyurishte) that our optician has managed to find in the drawer of his desk. In the preliminary experiments, we studied dynamics of photochromic darkening of the glass at different power densities of the irradiation. The experiments were performed in the pump-probe configuration using two light sources - a halogen incandescent lamp (P = 90W for the current 7.5A) as the pump and a laser diode (P ∼ 1 mW, λ ∼650 nm) as a probe (Fig. 2). The pump light was focused onto the glass as a spot ∼3x3 mm² in size. The probe laser beam passed through the sample practically in the opposite direction and initially was centered on the spot of the pump and then, when measuring, was translationally modulated at an audio frequency with the aid of a prism fastened to membrane of a loudspeaker (Fig. 2). Cross section of the laser beam (∼3 mm²) was substantially smaller than the size of the
spot, while the amplitude of its spatial displacement upon modulation was slightly larger than the spot. As a result, due to the photoinduced darkening of the glass, the intensity of the probe beam transmitted through the sample revealed modulation, proportional to the darkening, synchronized with vibrations of the membrane. The above modulation signal was lock-in amplified and recorded as a function of time. The photodetector was placed in the focal plane of the lens which allowed us to eliminate the parasitic modulation caused by spatial modulation of the spot over the photodetector. The current of the incandescent lamp was controlled by a special generator with the lower frequency bound of 0.001 Hz. Upon stepwise change of the pump light intensity, the time-dependent signal of the probe beam showed dynamics of recovery of the glass transmission. These measurements were performed under conditions of square-wave modulation of the lamp current at frequencies of about a few hundredths of Hz with the degree of intensity modulation of about 20 - 30 %. We have found that the relaxation dynamics strongly depended on the irradiation intensity. At low levels of illumination, the relaxation times were rather long (hundreds of seconds), then, as the pump intensity increased, they shortened to tens of seconds and then the situation became unstable: both the relaxation time and even the sign of the photochromism showed a long-term drift in the range of tens of minutes. The latter region was highly inconvenient for our measurements, and we have chosen the range of power densities where the relaxation times were relatively short. Figure 3 shows experimental time dependence of transmission of the glass sample upon square-wave modulation of the lamp current in the vicinity of 4.5 A. The relaxation time of the induced absorption, under these conditions, was around 10 s, though, strictly speaking, the relaxation process was not single-exponential.

The experiments were performed in a standard way, as in most experiments on “slow” and ”fast” light. Intensity of the pump light incident on the sample was subjected to a weak harmonic modulation, whose amplitude and phase were measured at the exit of the sample at different modulation frequencies. The results of the measurements are shown in Fig. 4. As expected for the inverse saturable absorber, the amplitude spectrum of the signal exhibited a dip in the range of low frequencies (rather than a peak typical for a bleachable absorber), while the phase spectrum corresponded to advancement of the output signal with respect to that at the entrance. In view of the non-exponential character of the relaxation mentioned above, the shape of the experimental frequency dependences did not exactly correspond to predictions of the simplest model [1], which, for our purposes, was of no importance. Figure
5 shows schematically mutual position of normalized curves of the light modulation at the entrance and at the exit of the sample for the modulation frequency 0.001 Hz. Temporal advancement of the output light was, in this case, $\sim 3$ s. If we assume, as it is accepted in the experiments on "slow" and "fast" light, that the observed phase delay is related to a change of the light group velocity in the medium, then we will have to acknowledge that the group velocity, in this case, is negative and equals $-0.67$ mm/s. Now, however, the possibility of such an interpretation is excluded, because there is no preferential frequency in the optical spectrum of the light source near which the appropriate anti-hole (with a width of about 0.1 Hz) is supposed to be burnt, and therefore the delay cannot be ascribed to a change in the group velocity of light in the sense of Eq. (4). It is evident that the terms "superluminal light" or "the light with negative group velocity" are also inappropriate here.

III. "ULTRASLOW LIGHT" IN A THERMOCHROMIC COATING

As the second object for study, we used a fragment of a cup with a thermochromic decorative coating that changed its color upon heating (in this particular case, the initially red coating was noticeably bleached upon heating to $40^\circ$ - $50^\circ$). Dynamics of response of the sample to variations of the light intensity was studied using the same incandescent lamp with the same experimental arrangement, with the only difference that now we detected the light scattered by the surface of the sample (rather than transmitted light). The photodetector was placed approximately at the angle of specular reflection, where the light intensity scattered by the glossy surface of the coating was the greatest. In front of the detector, we placed an optical filter transmitting blue-green part of the visible spectrum, which provided a higher contrast of the detected intensity nonlinearity. With increasing intensity of the incident light, the temperature of the sample was growing, the coating was getting bleached, and the effective reflectivity increased. Figure 6 shows the experimental dependence of equilibrium intensity of the reflected light on that of the incident light. The dependence was recorded under conditions of ultimately slow scanning of the lamp current (during 10 min). This is the basic dependence describing the intensity-type nonlinearity of the nonlinear medium [see Eq. (1)], which is, in this case, a saturable reflector. The growing reflectivity with increasing light intensity corresponded to the positive sign of the coefficient $K_1$ in Eq. (1). For such a high level of nonlinearity, dynamics of the relaxation behavior of
the sample could be easily observed directly in the reflected light upon square-wave modulation of the incident light intensity (Fig. 7). The recovery time of the sample, in our experimental conditions, lied in the range of 15 s. The experimental frequency dependence of the amplitude and phase of the reflected light intensity modulation are shown in Fig. 8. For normalization of the amplitude and phase of the reflected signal, we used an additional photodetector illuminated by the light split from the incident beam. The frequency behavior of the response, in this case, qualitatively coincided with what is usually observed for a bleachable absorber under conditions of laser excitation. The modulation amplitude shows a growth in the vicinity of zero frequencies (Fig. 8a), while the phase shift (Fig. 8b) reveals a peak in the vicinity of the frequency approximately corresponding to the inverse recovery time of the nonlinear medium, and, by its sign, to positive delay of the modulation with respect to that of the incident light. The value of this delay, in the limit of low frequencies, was about 2.5 s. In this case, however, this delay not only cannot be ascribed to the group velocity reduction, but, in addition, the group velocity cannot be estimated in a sensible way, though the experiment evidently may be performed, with the same results, using laser source

IV. CONCLUSIONS

In this paper, we have shown experimentally that the effects of the light pulse delay (or phase delay of the light intensity modulation) are inevitable in any medium with the intensity-type nonlinearity and, by themselves, do not evidence in favor of any changes in the light group velocity.

The essence of all the effects of this kind is reduced to changes in intensity spectrum of the light interacting with the nonlinear medium and, correspondingly, to changes in temporal behavior of its intensity. In the particular case of a sufficiently long pulse (when the main part of its intensity spectrum falls into the region of linear dependence of the intensity modulation phase on modulation frequency), the pulse distortion is reduced to its pure shift in time. In this case, the spectral composition of the optical field is important only to the extent of its influence on parameters of Eqs. (1) - (3).

For this reason, all the experimental results on the delay or advancement of the light
pulse propagating through a saturable absorber (starting from the paper [6]) are natural for the media with intensity nonlinearity and do not require, for their interpretation, invoking the effects of the light group velocity changes. In principle, the described variations in the intensity spectrum of the light interacting with a saturable medium should also be revealed in fine changes of the phases and amplitudes of the optical spectral components (this is, in fact, the essence of the CPO effect). However, the contribution of such processes into changes of the light group velocity can be revealed only in purposely designed experiments, which, to the best of our knowledge, so far has not been performed. Even in the experimental work [13], where the steep refractive index dispersion under conditions of the CPO effect was convincingly demonstrated, no direct measurements of the probe pulse delay in the presence of the pump have been made and compared with calculations. Such measurements make sense for two reasons. First, the CPO-related dip in the absorption spectrum is not the same for monochromatic probe and for the probe with a continuous spectrum (because of interaction between the spectral components of the probe symmetric with respect to the pump). In particular, as was pointed in [1], for the probe light with certain phase correlations between its ”mirror” components, the dip can be unobservable. And, second, it is known that the pulse delay can be also observed for the probe pulse wavelength shifted far away from that of the pump [14]. This is why it would be important to evaluate the ”resonant” CPO-based contribution to the delay of optical pulse in a saturable absorber.

In any case, we are not aware of any experimental observations on the ”CPO-based slow light” that cannot be interpreted in the framework of the simplest model of intensity nonlinearity. The same conclusion was made in [10] on the basis of a comprehensive analysis of many published experiments. In spite of the fact that the ”CPO-based slow light” is nowadays one of the basic technologies of ”slow light”, in our opinion, there is no evidence of its real experimental observation and no grounds to consider its existence proven.
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FIGURE CAPTIONS

FIG.1 Regularities of optical response of the medium with the intensity-type nonlinearity. a - response to a stepwise change of the incident light intensity; b and c - frequency dependences of the amplitude and phase of a sine-modulated beam at the exit of the medium; d - temporal shift of a smooth pulse of intensity modulation. Curves 1 and 2 correspond to the cases of superlinear and sublinear dependence (1).

FIG.2 Experimental setup for measuring dynamics of darkening in the photochromic glass. 1 - incandescent lamp, 2 - focusing lenses, 3 - photochromic glass, 4 - photodetectors, 5 - laser diod, 6 - vibrating prism, 7 - data processing system, 8 - power supply, 9 - controlling low-frequency generator.

FIG.3 Experimental time dependence of the photochromic glass darkening (a) induced by the square-wave modulated pump light (b).

FIG.4 Frequency dependences of the amplitude (a) and phase (b) of the sine-modulated light transmitted through the photochromic glass.

FIG.5 A sketch of time advancement of the sine-modulated light transmitted through the photochromic glass.

FIG.6 Experimental dependence $I_{out}(I_{in})$ for the thermochromic coating.

FIG.7 Time dependence of intensity of the light reflected from the thermochromic coating for the square-wave modulated incident light of the incandescent lamp.

FIG.8 Frequency dependences of the amplitude (a) and phase (b) of the sine-modulated light reflected from the thermochromic coating.
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