Slow light in saturable absorbers: Progress in the resolution of a controversy

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There are two opposing models in the analysis of the slow transmission of light pulses through saturable absorbers. The canonical incoherent bleaching model simply explains the slow transmission by combined effects of saturation and of noninstantaneous response of the medium resulting in absorption of the front part of the incident pulse larger than that of its rear. The second model, referred to as the coherent-population-oscillations (CPO) model, considers light beams whose intensity is slightly pulse modulated and attributes the time delay of the transmitted pulse to a reduction of the group velocity. We point out some inconsistencies in the CPO model and show that the two models lie in reality on the same hypotheses, the equations derived in the duly rectified CPO model being local expressions of the integral equations obtained in the incoherent bleaching model. When intense pulses without background are used, the CPO model, based on linearized equations, breaks down. The incoherent bleaching model then predicts that the transmitted light should vanish when the intensity of the incident light is strictly zero. This point is confirmed by the experiments that we have performed on ruby with square-wave incident pulses and we show that the whole shape of the observed pulses agrees with that derived analytically by means of the incoherent bleaching model. We also determine in this model the corresponding evolution of the fluorescence light, which seems to have been evidenced in other experiments.

I. INTRODUCTION

The group velocity is a basic concept in the study of the propagation of *coherent* light pulses of slowly varying amplitude in a *linear*, time-independent, dispersive medium. It is invariably introduced in every review on slow and fast light (see, e.g., [1,2]). Expressing that the phase of the optical field is stationary at the frequency ω_c of the pulse carrier, it is given without any ambiguity by the relation:

$$v_g(\omega_c) = \frac{c}{n(\omega_c) + \omega_c \, dn/d\omega_c}, \qquad (1)$$

where c is the light velocity in vacuum, $n(\omega)$ designates the refractive index at the optical frequency ω, and $dn/d\omega_c$ is a short-hand notation of its derivative for $\omega = \omega_c$. Slow light is obtained when $dn/d\omega_c$ takes large positive values (steep dispersion), the group velocity being then much smaller than the corresponding phase velocity $c/n(\omega_c)$. As a consequence of the Kramers-Kronig relations, this occurs in particular when the carrier frequency ω_c of the light pulses is close to the frequency ω_0 of a well-marked maximum of the medium transmission (minimum of absorption). When $\omega_c = \omega_0$, the group velocity is minimal and a simple application of the moment theorem shows that the center of gravity of the pulse envelope then exactly propagates at the corresponding group velocity whatever the pulse shape is [3]. It should be mentioned that, due to the unavoidable distortion of a pulse propagating in a dispersive absorptive medium, the location τ_{max} of its maximum generally differs from that τ_g of its center of gravity. It has been, however, demonstrated that, under certain general conditions, the transmitted pulse becomes nearly Gaussian for large enough propagation distances, with obviously $\tau_{max} \approx \tau_g$ [4].

An important issue for eventual applications of slow light is the fractional delay τ_{max}/τ_{in}, where τ_{in} is the full width at half-maximum (FWHM) of the intensity profile of the incident pulse. Large fractional delays (up to 25 with moderate pulse attenuation) have been evidenced when the required peak in the medium transmission is associated with the minimum of absorption occurring halfway between two absorption lines [5–7]. Most often, the transmission peak is obtained by applying an extra coherent wave interacting nonlinearly with the medium, exploiting, for instance, electromagnetically induced transparency (EIT) in atomic vapors [8] or Brillouin-induced gain in optical fibers [9]. Slow light becomes a fashionable topic with the demonstration in an EIT experiment of a group velocity as slow as 17 m/s in an ultracold atomic gas [10]. Subsequently the experiments showing delays in the transmission of light pulses were systematically analyzed in terms of slow group velocity. Such an analysis, indisputable for the EIT experiments, is, however, questionable in the cases where the group velocity as given by Eq. (1) is not well defined [11].

We specifically examine here the case of the transmission of light pulses through saturable absorbers. As far back as 1965, Gires and Combaud [12] showed that the stationary transmission of organic dyes is fairly well reproduced by assimilating the medium to a resonant two-level medium and using the rate equations approximation. By this means they obtained two equations coupling the population difference and the beam intensity. Solving these equations in the time-dependent case, Selden theoretically studied the transmission of light pulses, demonstrating narrowing, skewing, and time delay of the transmitted pulse [13,14] in agreement with the experimental observations [15–17]. From a qualitative viewpoint, the delay is thus interpreted in terms of *pulse reshaping*, the leading edge of the incident pulse being more attenuated than its trailing edge. Insofar as only the intensity is

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involved in the process, the phenomenon is currently referred to as *incoherent bleaching* to distinguish it from the pulse delay and reshaping that occur in purely coherent cases. Smith and Allen showed that delays sometimes attributed to self-induced transparency (a *coherent* phenomenon) are in fact the result of *incoherent* bleaching and are well reproduced by Selden's theory [18]. For a complete analysis of this point, see [19]. Always by using the incoherent bleaching model, Selden also examined the transmission of a continuous wave (cw) whose intensity is slightly modulated by a sine wave [20]. He showed that the intensity modulation index increases with the propagation distance whereas its phase is time delayed. These phenomena were observed on a ruby crystal at room temperature by Hillmann *et al.* [21] and by Bigelow *et al.* [22]. The latter, not taking into account the results of the incoherent bleaching model and considering the time delay of the modulation phase to be a group delay, claimed to have discovered "a new method that produces slow propagation of light" [22]. Their theoretical analysis was based on an extrapolation of the results obtained when two separated *coherent* waves originate *coherent* oscillations of the populations at their frequency difference. Abundantly cited, Ref. [22] paved the way to numerous articles invoking slow light based on coherent population oscillations (CPO). The systems under consideration are very various, comprising in particular doped crystals [23–25], semiconductor devices [26–30], doped optical fibers [31–34], and doped glass microspheres [35]. However, as shown in [11,36–38], the effects reported in most of these articles do not involve coherence in the optical sense and can be explained in the frame of the incoherent bleaching model. The controversy on this matter restarted more recently with the publication of an article reporting experiments performed with a spinning ruby window [39]. In this article Wisniewski-Barker *et al.* claim that their results are "incompatible with slow-light models based on simple pulse reshaping arising from optical bleaching." This statement was contested by Kozlov *et al.* [40] who performed an experiment validating the incoherent bleaching model but, surprisingly enough, Wisniewski-Barker *et al.* obtained the opposite result by using practically the same experimental setup [41]. The controversy between the two models thus remains open. We attempt in the following to solve this issue. In Sec. II, we revisit the case of weak modulation depths by extending the results given in [20,37] and showing that, after correction, the CPO equations are simply local expressions of the integral equations obtained in the incoherent bleaching model. In Sec. III, we examine the validity of the bleaching model in the case of saturating pulses without background, we report experiments confirming the positive result of Kozlov *et al.* and we give a possible explanation of the different result reported in [41]. We finally conclude in Sec. IV by summarizing and discussing our main results.

II. CASE OF WEAK MODULATION DEPTHS

The CPO model traces back to the paper of Schwarz and Tan who studied how the absorption of a coherent probe wave by a saturable absorber is modified when the medium is submitted to a coherent saturating wave [42]. They showed

that the probe absorption spectrum then displays a dip (hole) centered at the pump frequency and of width $\approx 1/T_1$, where T_1 is the population relaxation time. This dip is considered in [21] as resulting from the population oscillations created in the medium by the beating of the pump and probe waves. When the directions of propagation of the two waves are different, it is possible to determine without ambiguity the absorption coefficient, the refractive index, and the group velocity of the probe wave. An experiment corresponding to this scheme has been performed by Ku *et al.*, the slow group velocity being inferred from a measurement of the phase of the probe field [26]. A related experiment involving counter-propagative waves is reported in [34].

The problems arise when the previous results are extended to the study of a single cw whose intensity is slightly modulated by a sine wave of low frequency. Denoting Ω the modulation frequency, the modulated wave can be considered as the superimposition of three co-propagative cws, a saturating wave of frequency ω_s and two sidebands of frequency $\omega_s \pm \Omega$, acting as probes [43]. Bigelow *et al.* consider in [22] that the two probes act independently but, as soundly noted by Sargent, "although neither probe frequency could influence the other on its own, they succeed in doing so with the help of the saturating wave" [44]. This indicates that Eq. (9) in [22] is not correct. This error has been pointed out for the first time by Mørk *et al.* [28] who indicated that a correct application of the four-wave mixing theory leads to multiply the phase lag of the modulation and thus the corresponding time delay by a factor two.

Another point raised by Zapasskii and Kozlov [36] is that the CPO model implicitly assumes that the saturating wave has a spectral width much smaller than that of the hole induced in the absorption spectrum ($\approx 1/T_1$). This condition is far from being met in most of the experiments. There is then no dip in the *optical* absorption spectrum and thus no associated slow light in the usual sense. In response to this objection, Piredda and Boyd [45], while continuing to invoke CPO, developed a model based on two equations coupling wave intensity and ground-state population identical to those coupling wave intensity and population difference in the incoherent bleaching model [12,13,37]. They can be written in the simplified form,

$$T_1 \frac{\partial N}{\partial t} = -N(1 + I) + 1, \tag{2}$$

$$\frac{\partial I}{\partial z} = -\alpha I N, \tag{3}$$

where T_1 is the ground-state recovery time, N is the ground-state population normalized to its value at equilibrium, t is the time retarded by the propagation time in the host medium (negligible compared to the delays considered in the following), I is the beam intensity normalized to the saturation intensity, z is the abscissa along the direction of propagation, and α is the unsaturated absorption coefficient. Equations (2) and (3) implicitly assume that the ground state has no resolved structure. This condition is met in saturable absorbers which are dense media working at room temperature. It prevents oscillations at the frequency of a transition between two separated sublevels as evidenced in an alkali-metal vapor in

buffer gas [46,47]. In the case of a cw, Eqs. (2) and (3) are reduced to

$$\bar{N}(z) = \frac{1}{1 + \bar{I}(z)}, \tag{4}$$

$$\frac{\partial \bar{I}(z)}{\partial z} = -\frac{\alpha \bar{I}(z)}{1 + \bar{I}(z)}, \tag{5}$$

$$\bar{I}(z) + \ln \bar{I}(z) = \bar{I}(0) + \ln \bar{I}(0) - \alpha z, \tag{6}$$

where, as in the following, upper bars refer to time-independent quantities. When the cw is slightly modulated, $I(z,t) = \bar{I}(z) + \Delta I(z,t)$ with $\Delta I(z,t) \ll \bar{I}(z)$ and $N(z,t) = \bar{N}(z) - \Delta N(z,t)$ with $\Delta N(z,t) \ll \bar{N}(z)$. Making a calculation at the first order in ΔI and ΔN, taking into account Eqs. (4) and (6) and passing in the Fourier space [48], we obtain the transfer functions $H(z,\Omega)$ relating the Fourier transform $\Delta I(z,\Omega)$ of $\Delta I(z,t)$ to that $\Delta I(0,\Omega)$ of $\Delta I(0,t)$ and $H_{\Delta N}(z,\Omega)$ relating the Fourier transform $\Delta N(z,\Omega)$ of $\Delta N(z,t)$ to $\Delta I(z,\Omega)$. They read

$$H(z,\Omega) = \left(\frac{\bar{I}(z)}{\bar{I}(0)} \right) \left(\frac{1 + \bar{I}(0) + i\Omega T_1}{1 + \bar{I}(z) + i\Omega T_1} \right)$$
$$= \left(\frac{\bar{I}(z)}{\bar{I}(0)} \right) \left(1 + \frac{\bar{I}(0) - \bar{I}(z)}{1 + \bar{I}(z) + i\Omega T_1} \right). \tag{7}$$

$$H_{\Delta N}(z,\Omega) = \frac{1}{[1 + \bar{I}(z)][1 + \bar{I}(z) + i\Omega T_1]}. \tag{8}$$

The transfer function $H(z,\Omega)$ has a single pole and a single zero, both purely imaginary with a positive imaginary part. This implies that its inverse Fourier transform, that is, the system impulse response, is real and that the system is causal with minimum phase shift [48]. The phase $\Phi(z,\Omega)$ of $H(z,\Omega)$ then obeys to the relation,

$$\Phi(z,\Omega) = -\mathcal{H}\{|\ln(H(z,\Omega))|\}, \tag{9}$$

where $\mathcal{H}$ designates the Hilbert transform. Equation (9) may be considered as a generalized Kramers-Kronig relation. Similar properties hold for $H_{\Delta N}(z,\Omega)$. We incidentally remark that $H(z,\Omega)$ is the transfer function of a simple electric network involving an RC circuit, two voltage dividers, and a voltage adder whereas an RC circuit and one voltage divider suffice to reproduce $H_{\Delta N}(z,\Omega)$.

To relate Eq. (7) to the results given by Piredda and Boyd in [45], we consider the transfer function $H(dz,\Omega) = H(z + dz,\Omega)/H(z,\Omega)$ of the infinitely thin slice comprised between z and $z + dz$ in the medium. Taking into account Eq. (5), we obtain

$$H(dz,\Omega) = \exp\left[-\alpha_{\mathrm{mod}}(z,\Omega)dz - i\varphi_{\mathrm{mod}}(z,\Omega)dz\right], \tag{10}$$

with

$$\alpha_{\mathrm{mod}}(z,\Omega) = \frac{\alpha}{1 + \bar{I}(z)} \left\{ 1 - \frac{\bar{I}(z)[1 + \bar{I}(z)]}{[1 + \bar{I}(z)]^2 + (\Omega T_1)^2} \right\}, \tag{11}$$

$$\varphi_{\mathrm{mod}}(z,\Omega) = \frac{\alpha \bar{I}(z)}{1 + \bar{I}(z)} \left\{ \frac{\Omega T_1}{[1 + \bar{I}(z)]^2 + (\Omega T_1)^2} \right\}. \tag{12}$$

In relation with Eq. (9), we note that $\varphi_{\mathrm{mod}}(z,\Omega) = \mathcal{H}[\alpha_{\mathrm{mod}}(z,\Omega)]$. When the modulation is reduced to a sine wave of frequency Ω (as considered in most experiments), $\alpha_{\mathrm{mod}}(z,\Omega)$ is the attenuation coefficient of the modulation, and

$\varphi_{\mathrm{mod}}(z,\Omega)$ is the associated phase lag per length unit. Equations (11) and (12) are strictly equivalent to those obtained in [45] by invoking the CPO model and this shows that *the CPO model does not bring new results with regard to the incoherent bleaching model*. As expected, the phase lag is two times that given in [22,49]. A phase velocity $v_{\varphi\mathrm{mod}}(z,\Omega) = \Omega/\varphi_{\mathrm{mod}}(z,\Omega)$ can be associated with this phase lag. Depending on Ω, *this velocity should not be confused with a group velocity* as it is made in [22,45]. When the modulation consists in a pulse, the modulation group delay $d\tau_{\mathrm{gmod}}$ through the slice $(z,z + dz)$ is derived from $H(dz,\Omega)$ by the moment theorem. The slice being assumed to be infinitely thin $H(dz,0) \approx 1$ and $d\tau_{\mathrm{gmod}}$ can be identified to the coefficient of the first degree term in the expansion of $H(dz,\Omega)$ in power series of $(-i\Omega)$. We get

$$d\tau_{\mathrm{gmod}} = \frac{\alpha \bar{I}(z)T_1 dz}{[1 + \bar{I}(z)]^3}, \tag{13}$$

and the corresponding (local) group velocity $v_{\mathrm{gmod}}(z) = dz/d\tau_{\mathrm{gmod}}$ reads

$$v_{\mathrm{gmod}}(z) = \frac{[1 + \bar{I}(z)]^3}{\alpha \bar{I}(z)T_1}. \tag{14}$$

Note that this group velocity is related to the intensity modulation transmission and should be distinguished from the group velocity as defined by Eq. (1) for pulses of coherent light.

The attenuation of the cw and of the modulation being intrinsically coupled, the widespread approximation consisting in neglecting the former to study the latter is not justified. The medium being assumed to occupy the space $0 \leqslant z \leqslant L$, the use of the integral expressions of Eqs. (7) and (8) has the advantage to give directly $\Delta I(L,\Omega)$ and $\Delta N(L,\Omega)$ without requiring integration in z. The corresponding expressions of $\Delta I(L,t)$ and $\Delta N(L,t)$ read

$$\Delta I(L,t) = \mathcal{F}^{-1}[H(L,\Omega)\Delta I(0,\Omega)], \tag{15}$$

$$\Delta N(L,t) = \mathcal{F}^{-1}[H_{\Delta N}(L,\Omega)H(L,\Omega)\Delta I(0,\Omega)], \tag{16}$$

where $\mathcal{F}^{-1}$ designates inverse Fourier transforms. General characteristics of $\Delta I(L,t)$ and $\Delta N(L,t)$ can be derived by exploiting the remarkable properties of the cumulants [4,50]. The cumulants κ_n of the Fourier transform $G(\Omega)$ of a real function $g(t)$ are given by the expansion,

$$G(\Omega) = G(0)\exp\left(\sum_{n=1}^{\infty} \frac{\kappa_n}{n!}(-i\Omega)^n\right), \tag{17}$$

where the cumulants κ_1, κ_2, and κ_3 can be shown to be, respectively, equal to the mean time of $g(t)$, its variance σ^2, and its third centered moment μ_3. For $G(\Omega) = H(L,\Omega)$, we get $\kappa_1 = T_L - T_0$, $\kappa_2 = T_L^2 - T_0^2$, and $\kappa_3 = 2(T_L^3 - T_0^3)$ where T_z is a short-hand notation of $T_1/[1 + \bar{I}(z)]$. When the modulation is pulsed, the additivity property of the cumulants enables us to identify κ_1 to the time delay of the pulse center-of-gravity (modulation group delay), κ_2 to the increase of the pulse variance, and κ_3 to that of μ_3. The group delay for the whole medium thus reads

$$\tau_{\mathrm{gmod}}(L) = \frac{T_1}{1 + \bar{I}(L)} - \frac{T_1}{1 + \bar{I}(0)}. \tag{18}$$

We incidentally remark that this result can be retrieved by an integration combining Eqs. (13) and (5). An important point is that the group delay cannot exceed T_1 as large as the medium thickness may be. This limit is attained when $\bar{I}(0) \gg 1$ (strongly saturating incident cw) and $\bar{I}(L) \ll 1$ (medium nearly opaque). The variance and the third moment of the transmitted pulse are then also the largest. If the incident pulse is symmetric, the large positive value of κ_3 entails that the output pulse will be strongly skew with a rise much steeper than its fall and, consequently, a time-delay $\tau_{\max}$ of its maximum much shorter than the group delay τ_{gmod}. See, for illustration, Fig. 2(c) in [37]. In the more realistic case where $\bar{I}(0) < 1$ [always with $\bar{I}(L) \ll 1$], the pulse distortion will be moderate but the group delay will be short compared to T_1 (while keeping significantly longer than $\tau_{\max}$). When the incident pulse is a Gaussian of the form $\Delta I(0,t) \propto \exp\left(-t^2/\tau_p^2\right)$, the transmitted pulse derived from Eq. (15) reads

$$\Delta I(L,t) \propto \Delta I(0,t) + \frac{\bar{I}(0)\tau_p\sqrt{\pi}}{2T_1}$$
$$\times \left[1+\mathrm{erf}\left(\frac{t}{\tau_p}-\frac{\tau_p}{2T_1}\right)\right]\exp\left(\frac{\tau_p^2}{4T_1^2}-\frac{t}{T_1}\right),$$
(19)

where erf designates the error function. Figure 1 shows the result obtained in the conditions of the experiment on ruby reported in [25]. The parameters are $\bar{I}(0) = 0.23$, $T_1 = 1.6$ ms, $\alpha = 1.17\,\mathrm{cm}^{-1}$, and $L = 4.25$ cm, from which we deduce $\alpha L \approx 5$, $\bar{I}(L) \approx 2\times10^{-3}$, $T_L \approx T_1$, and $\tau_{\mathrm{gmod}}(L) \approx 0.3$ ms (actually much shorter than T_1).

The incident pulse (dotted line) has an FWHM duration $\tau_{\mathrm{in}} = 2\tau_p\sqrt{\ln 2} = 5$ ms. As predicted, $\tau_{\max}$ is significantly shorter than τ_{gmod} ($\tau_{\max} \approx \tau_{\mathrm{gmod}}/2$). We also compare in Fig. 1 $\Delta N(z,L)$ to $\Delta I(z,L)$. It is easily deduced from Eq. (8) that the group delay of $\Delta N(z,L)$ exceeds that of $\Delta I(z,L)$ by a
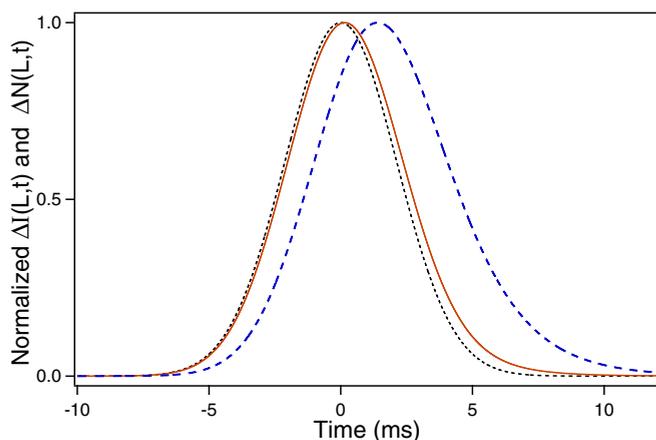

FIG. 1. Normalized transmitted pulse $\Delta I(L,t)$ (solid line) and population variation $\Delta N(L,t)$ (dashed line). Parameters are as follows: $\bar{I}(0) = 0.23$, $T_1 = 1.6$ ms, $\alpha = 1.17\,\mathrm{cm}^{-1}$, and $L = 4.25$ cm, leading to $\alpha L \approx 5$, $\bar{I}(L) \approx 2\times10^{-3}$, $T_L \approx T_1$, $\Delta\tau_g(L) \approx T_1$, and $\tau_{\mathrm{gmod}}(L) \approx 0.3$ ms. The normalized incident pulse $\Delta I(0,t)$ (dotted line) of FWHM duration $\tau_{\mathrm{in}} = 5$ ms is given for reference. Note the large delay of $\Delta N(L,t)$ compared to that of $\Delta I(L,t)$.
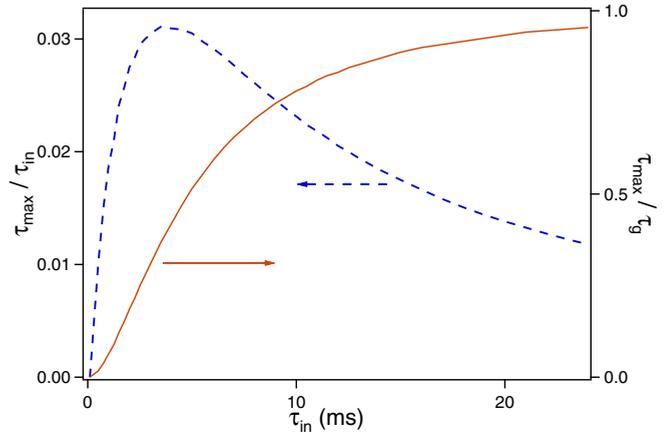

FIG. 2. Fractional delay $\tau_{\max}/\tau_{\mathrm{in}}$ of the transmitted pulse (dashed line, left scale) and ratio $\tau_{\max}/\tau_{\mathrm{gmod}}$ (solid line, right scale) as functions of the duration τ_{in} of the incident pulse. Other parameters as in Fig. 1.

quantity,

$$\Delta\tau_{g\Delta N} = \frac{T_1}{1 + \bar{I}(L)} \approx T_1. \tag{20}$$

An important point is that this extra delay of the population evolution is much longer than the delay of the signal $\Delta I(z,L)$.

Coming back to the latter, Fig. 2 shows how the fractional delay $\tau_{\max}/\tau_{\mathrm{in}}$ of the transmitted pulse and the ratio $\tau_{\max}/\tau_{\mathrm{gmod}}$ depend on the duration τ_{in} of the incident pulse. It appears that $\tau_{\max}$ approaches its asymptotic value τ_{gmod} for values of τ_{in} at which the fractional delay tends to 0 and, conversely, that the latter attains its maximum for a pulse duration τ_{in} such that is only about $\tau_{\mathrm{gmod}}/3$.

III. CASE OF INTENSE PULSES WITHOUT BACKGROUND

Even duly rectified, the CPO model, based on linearized equations, does not apply to the case of saturating pulses without background. In order to extend its range of application to such situations, its defenders invoke a mechanism of "self-pumping," one part of the pulse acting as a pump whereas the remaining part acts as the probe [22,41]. Without any quantitative support, this claim seems purely incantatory. It is even qualitatively incompatible with the fact that smooth symmetric pulses are broadened and gain a positive skewness (rise steeper than the fall) when they are superimposed on a large background (range of validity of the CPO model) whereas saturating pulses without background are narrowed and gain a negative skewness (fall steeper than the rise) [13,15,37]. Reporting experiments performed with a spinning ruby window, Wisniewski-Barker *et al.* [39] recently proclaimed the failure of the incoherent bleaching model for this reference material. According to the demonstration made in the previous section of the equivalence of the two models, all the results obtained by means of the CPO model in the weak modulation limit would then require revision. Fortunately enough, it is nothing of the sort. The argument given in [39] against the incoherent bleaching model is that, in this model, the transmitted light intensity should vanish when the incident light intensity is
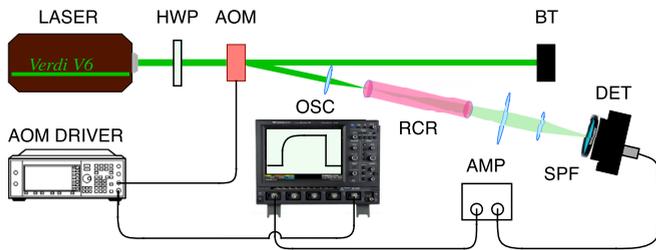
FIG. 3. Experimental setup. LASER, single longitudinal mode Nd:YVO$_4$ laser (Verdi V6, Coherent) delivering a cw at 532 nm; HWP, half-wave plate; AOM, acousto-optic modulator (AA Opto-Electronic, MTS110-A-VIS); BT, beam trap absorbing the non-diffracted light; RCR, ruby crystal rod; SPF, dichroic shortpass filter (Semrock, BSP-633R-25) eliminating the ruby fluorescence light at 694 nm; DET, high-speed silicon photodetector (Thorlabs DET 210, DC-350 MHz); AMP, low-band (DC-200 kHz) amplifier (Hamamatsu C7319); OSC, digital oscilloscope (Lecroy Waverunner 104 MXi); AOM DRIVER, function generator driving the acousto-optic modulator and triggering the oscilloscope.

FIG. 4. Example of transmitted signal observed in our experiments for a power sent on the ruby crystal of about 1.6 W and a pulse duration $\tau_{\mathrm{in}} = 25$ ms (solid line). The dashed line is the profile of the incident pulse obtained by replacing the ruby crystal by a suitable neutral density filter. The insets are enlargements of the pulse rise and fall.

null, a condition that would be not verified in the experiments. In a comment, Kozlov *et al.* [40] contested the achievement of strictly null incident intensity in these experiments and proposed a more drastic test where the incident beam is switched on-off by a mechanical chopper (100% square wave modulation). Performing this experiment, they obtained results validating the incoherent bleaching model, without the slightest transmitted intensity after the switching off of the incident beam. This, however, did not close the debate. Indeed, accepting the challenge proposed by Kozlov *et al.*, Wisniewski-Barker *et al.* [41] performed a nearly identical experiment. They obtained opposite results, evidencing in particular an exponential decrease of the transmitted intensity after the switching off of the incident beam. Note that they did not attempt to explain the discrepancy between their results and those of Kozlov *et al.* To solve this issue, we describe in the following an experiment that validates the incoherent bleaching model and theoretically determine in this model the time dependence of $I(L,t)$ and of $N(L,t)$, providing a possible explanation of the experimental results obtained in [41].

Figure 3 shows our experimental setup. It is very similar to those used in [40,41]. We use a single longitudinal mode Nd:YVO$_4$ laser (Verdi V6, Coherent) operating at 532 nm as a cw source of controllable power. The laser beam is collimated and sent on an acousto-optic modulator (AA Opto-Electronic, MTS110-A-VIS). The light polarization at the input of the modulator is adjusted by a half-wave plate. A sine wave, at a 110-MHz acoustic frequency, is applied to the modulator crystal. Its amplitude is driven by a function generator which enables us to switch the power diffracted in the first order from 0% to 80% of the power delivered by the laser. The switching times are about 1 μs. A lens of 50-mm focal length focuses the diffracted beam slightly behind the front face of a 120-mm long standard laser ruby crystal rod. The transmitted beam is focused onto a high-speed silicon photodetector (Thorlabs DET 210, DC-350 MHz). The detector is preceded by a dichroic shortpass filter (Semrock, BSP-633R-25) of optical density exceeding 7 at 694 nm, which eliminates the fluorescence light emitted by the ruby crystal.
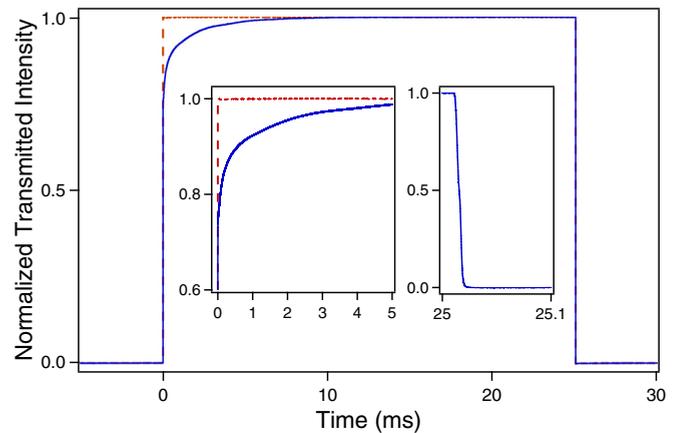
The signal delivered by the detector is amplified by a low-band (DC-200 kHz) amplifier (Hamamatsu C7319) and averaged by a digital oscilloscope (Lecroy Waverunner 104 MXi). We have carefully verified that the power diffracted in the first order is null when the modulator is in the off position (amplitude of the acoustic wave equal to zero) and that the corresponding signal delivered by the low-band amplifier vanishes. Numerous experiments have been performed for various laser powers and for several positions of the minimum beam waist inside the ruby crystal. In all these experiments, we never observe any detected signal when the power is switched off.

Figure 4 gives an example of detected signal obtained in such conditions. The power applied on the front face of the ruby crystal is modulated from 0 to 1.6 W by a 20-Hz square wave. The leading edge of the signal is characterized by an almost instantaneous jump which brings the signal to a value equal to 75% of its maximum value. This rapid variation is followed by a nearly exponential slow increase with a time constant of about 1.6 ms. In accordance with the incoherent bleaching model, on its trailing edge the signal quickly returns to zero with a 90%–10% switching time <6 μs mainly introduced by the amplifier. Note that, for completeness, we have also performed experiments where the acousto-optic modulator was replaced by a mechanical chopper as in [40,41]. The results obtained with both modulators are identical. Finally, although this parameter is not critical, we have measured the unsaturated optical thickness of the ruby crystal by collimating the beam inside the rod. The linear evolution of the transmitted power versus the input one leads to a mean value of αL close to 9.

To analyze the transmitted signals obtained in our experiments as those reported in [40,41], we come back to Eqs. (2) and (3) which may be considered as the basis of the incoherent bleaching model. They first show that $I(L,t) = 0$ when $I(0,t) = 0$. Analytical results valid at every time can be obtained when the medium thickness is such that $I(L,t) \ll I(0,t)$, a condition met in the experiments. As in [13,37], we
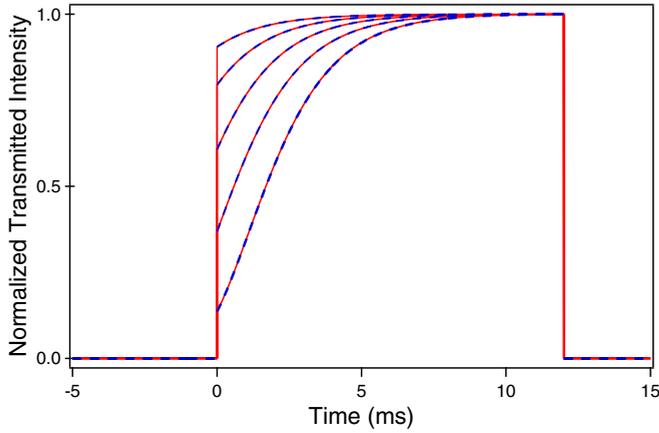
FIG. 5. Theoretical transmitted intensity as a function of time for an incident square pulse. Parameters are as follows: $T_1 = 1.6\,\mu$s, $\alpha L = 9$, $\tau_{in} = 12$ ms, and, from top to bottom, $I_0 = 0.1, 0.23, 0.5, 1$, and 2. The solid and dashed lines are, respectively, the exact numerical solution and the approximate analytic solution given by Eq. (24).

introduce the function,

$$Z(L,t) = \ln[I(L,t)] - \ln[I(0,t)] + \alpha L. \quad (21)$$

From Eq. (2,3), we deduce

$$T_1 \frac{dZ}{dt} + Z = I(0,t) - I(L,t) \approx I(0,t), \quad (22)$$

and

$$I(L,t) \approx I(0,t) \exp[Z(L,t) - \alpha L]. \quad (23)$$

In the case of a square incident pulse of the form $I(0,t) = I_0[u_H(t) - u_H(t - \tau_{in})]$ where $u_H(t)$ designates the Heaviside unit step function and τ_{in} is the pulse duration, we get

$$I(L,t) \approx I(0,t)\, e^{-\alpha L} \exp[I_0(1 - e^{-t/T_1})]. \quad (24)$$

The transmitted pulse displays an initial discontinuity equal to $I(0,t)\, e^{-\alpha L}$ at $t = 0$ before rising as an exponential of exponential and falling to 0 at $t = \tau_{in}$. If $\tau_{in} \gg T_1$, it attains the asymptotic limit $I_0 \exp(I_0 - \alpha L)$ before falling. Note also that the rise is reduced to a simple exponential if $I_0 \ll 1$. We have then

$$I(L,t) \approx I(0,t)\, e^{-\alpha L}[1 + I_0(1 - e^{-t/T_1})]. \quad (25)$$

Figure 5 shows the results obtained for $T_1 = 1.6\,\mu$s, $\alpha L = 9$, $\tau_{in} = 12$ ms, and, from top to bottom, $I_0 = 0.1, 0.23, 0.5, 1$, and 2. Note that the analytical solution given by Eq. (24) perfectly fits the exact numerical solution as long as $I_0 \ll \alpha L$ and that the shape of the transmitted pulse then does not depend on αL. The values $I_0 = 0.23$ and $T_1 = 1.6\,\mu$s approximately correspond to the experiment of Fig. 4 and Eq. (25) satisfactorily holds in this case.

In order to determine the normalized population $N_F \approx 1 - N$ of the fluorescent metastable level (again in the limit $I_0 \ll \alpha L$), we come back to Eq. (22) and replace $I(L,t)$ by its approximate form given by Eq. (24). After a tedious calculation, we get for the rise of $N_F(L,t)$ the rather complex expression,

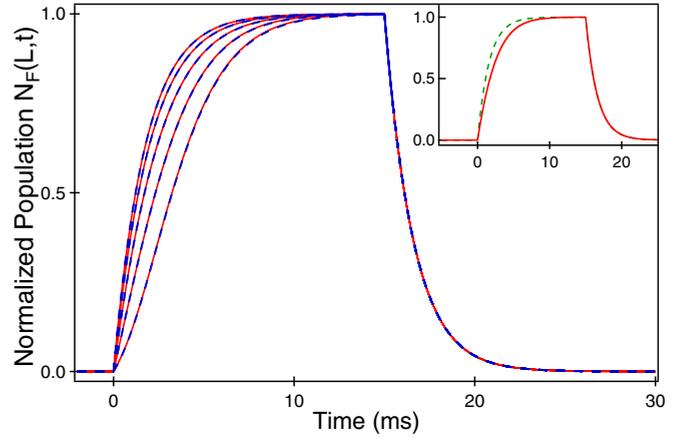$$N_F(L,t) = I_0 e^{-\alpha L} f(t)\, e^{-t/T_1} u_H(t), \quad (26)$$

FIG. 6. Theoretical evolution of the fluorescent metastable level population. Pulse duration $\tau_{in} = 15$ ms. Other parameters as in Fig. 5. The solid and dashed lines are, respectively, the exact numerical solution and the analytic solution given by Eqs. (26)–(28). (Inset) Comparison for $I_0 = 0.5$ of the exact numerical solution (solid line) to that given by Eqs. (28) and (29) analogous to those of an RC circuit (dashed line).

with

$$f(t) = \exp\left[\frac{t}{T_1} + I_0(1 - e^{-t/T_1})\right]$$
$$+ I_0 e^{I_0}[E_1(I_0) - E_1(I_0 e^{-t/T_1})] - 1, \quad (27)$$

where $E_1(x)$ designates the exponential integral function [51]. Equations (26) and (27) hold for arbitrary $I_0 \ll \alpha L$. For $t > \tau_{in}$, it immediately results from Eq. (3) that, without any restriction on I_0 and αL, $N_F(L,t)$ is reduced to a decreasing exponential of time constant T_1, namely

$$N_F(L,t) = N_F(L,\tau_{in}) \exp\left(-\frac{t - \tau_{in}}{T_1}\right) u_H(t - \tau_{in}). \quad (28)$$

Figure 6 shows the evolution of the population of the fluorescent metastable level obtained for the intensities already considered in Fig. 5. We see that Eqs. (26)–(28) perfectly fit the exact numerical solutions. When $I_0 \ll 1$, Eq. (26) becomes at the lowest order in I_0,

$$N_F(L,t) = I_0 e^{I_0 - \alpha L}[1 - e^{-t/T_1}]u_H(t). \quad (29)$$

Equations (29) and (28) are identical to those describing, respectively, the charge and the discharge of a capacity C through a resistance R with $RC = T_1$. As shown in the inset of Fig. 6, they provide a satisfactory approximation of the exact result for I_0 as large as 0.5.

Our experimental results on the transmitted pulse $I(L,t)$ obtained for an incident square pulse confirms those obtained by Kozlov *et al.* [40] and are in good agreement with our calculations based on the incoherent bleaching model. The key points are (i) $I(L,t)$ presents a discontinuity followed by an exponential-like rise when the incident beam is switched on (ii) $I(L,t)$ immediately falls to 0 when the incident beam is switched off. Quite different results are reported by Wisniewski *et al.* [41]: There is no initial discontinuity of $I(L,t)$ and $I(L,t)$ falls down exponentially with a time constant of about T_1 when the incident beam is switched off.

In these experiments, the signal hardly exceeds the dark signal of the detector and the signal-to-noise ratio is poor compared to that obtained by Kozlov *et al.* and in our experiments. The shape of the observed signals strangely resembles that of the evolution of the fluorescent metastable level population N_F obtained in the incoherent bleaching model (Fig. 6). It thus seems that the *light actually observed in* [41] *is nothing but fluorescence light*. The fact that "the delays of the individual Fourier components [of the observed signal] are independent of the modulation frequency" as noted in [41] simply reflects that, for moderate saturations, the population $N_F(L,t)$ practically evolves as the voltage in an RC circuit [see inset of Fig. 6 and Eqs. (28) and (29)]. Wisniewski *et al.* add "the shape of the tail should be independent of the modulation frequency of the pulse." Equation (28) shows that it is actually the case for $N_F(L,t)$ whatever the saturation is. Otherwise said, all the experimental results reported in [41] are quite compatible with the incoherent bleaching model insofar as the observed light seems to be fluorescence light and not the light transmitted at the laser frequency.

IV. SUMMARY AND DISCUSSION

Our article confirms the validity of the bleaching model to analyze the slow transmission of light pulses through saturable absorbers. Most experiments of so-called "Slow light based on (by means of, via) *coherent* population oscillations" are fully explained by the *incoherent* bleaching model. The very expression of coherent population oscillations is misleading. Indeed coherence in the optical sense generally plays no role in these experiments and the population oscillations at the low frequency of the intensity modulation are a trivial consequence of the equations coupling intensity and populations in saturable absorbers [12–14,20,37,45]. The population change being delayed with respect to the modulation (Fig. 1), the attribution of the origin of the slow transmission of the modulation to the population oscillations is questionable. We finally remark that the concept of group velocity as defined for pulses of coherent light does not apply to the broadband light considered in most experiments. It is, however, possible to define a group delay for the modulation [Eq. (18)] and even to establish a generalized Kramers-Kronig relation between its phase and its amplitude [Eq. (9)]. Note that these results are consistent with the CPO model as revised in [45] and that the equations obtained in the latter are only local expressions of the integral equations derived in the incoherent bleaching model. An important point is that the modulation group delay has an upper limit equal to the medium response time T_1 no matter the medium thickness and is often much shorter. See Figs. 1 and 2. This situation contrasts with that encountered in "pure" slow light

experiments performed with coherent light where the time delays do not suffer such limitations [52]. As soundly remarked in [38], it thus appears that the slow transmission in saturable absorbers reflects "slow response" of the medium rather than, strictly speaking, "slow light."

The above analysis essentially concerns the transmission of light pulses superimposed on a large background. For saturating pulses without background, the range of validity of the CPO model, based on linearized equations, is artificially extended in [22,41] by invoking a mechanism of self-pumping, one part of the pulse acting as a pump and the remaining part as a probe. Without any theoretical justification, this model gives pulse shapes that are qualitatively inconsistent with those derived in the incoherent bleaching model [13,14,37] and actually observed for organic dyes [15,16]. The recent claim of the failure of the incoherent bleaching model to explain the pulse shapes observed in the reference case of ruby at room temperature [39] originates two complementary experiments with this material [40,41]. Incident square wave pulses were used in these experiments, the main point being that, in the bleaching model, the transmitted light should immediately vanish at the instant where the incident light is switched off. The experiments were performed with similar setups but, surprisingly enough, gave opposite results. The first one [40] validates the bleaching model whereas the following one [41] shows an exponential-like fall of the transmitted pulse when the incident light is switched off. No explanation was given in [41] of this discrepancy. Our experiments and our theoretical calculations of the transmitted pulse shape and of the ruby fluorescent metastable state population bring some light on the problem. Our experimental result (Fig. 4) not only shows that the observed signal falls to 0 at the end of the square but that its rise is also in agreement with that predicted by the bleaching model (Fig. 5). The apparently opposite result obtained in [41] can be explained by examining the theoretical evolution of the population of the fluorescent state (Fig. 6) that strangely resembles the observed signal in [41]. We believe that the latter is caused by the fluorescence and the incoherent bleaching model is thus entirely validated.

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