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Kinks: Longitudinal excitation discontinuities in increasing absorption optical bistability

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Longitudinal excitation discontinuities (kinks) that arise from partial-sample switching in increasing absorption bistability are predicted theoretically and observed in a sharp-cut color filter. The kinks give rise to a sawtooth output temporal dependence for a triangular input shape. Kinks are an emphatic illustration of the local nature of increasing absorption bistability.

Koch, Schmidt, and Haug[1] have solved the transport equations for the light intensity and the excitation density for a system that exhibits increasing absorption[2–7] optical bistability. If the input light intensity is increased linearly at a rate appropriate for the excitation lifetime, a discontinuity in the excitation density occurs between the front of the sample, which has switched "off," and the back, which is still "on." This kink jumps discontinuously along the beam propagation direction. Consequently, the transmitted intensity has a sawtooth temporal dependence: The transmission drops each time the kink jumps deeper into the absorber. Herein is reported the first observation of such a kink, emphasizing the fact that increasing absorption bistability is a local phenomenon,[4,5] so that partial-sample switching is possible. Optical bistability[6,7] employing a cavity or other external feedback can have a longitudinal spatial dependence[8–10] due to finite absorption (breakdown of mean-field approximation), but a longitudinal discontinuity, i.e., partial-sample switching, is impossible. Spatial effects in optical bistability in cavities have been of considerable interest, but the emphasis has been upon transverse effects.[11] If the Fresnel number is large, a transverse discontinuity[12] in intracavity light intensity and medium polarization can occur, giving rise to transverse solitary waves[13,14] in the self-focusing case.

If the input intensity is an increasing ramp, the front part of the absorbing sample reaches the condition (carrier density, temperature, or whatever) for switching down before the back part whose input intensity is reduced by absorption in the front part.[1,15] By adjusting the rise time of the input, one can adjust the length of the front part that switches down. This can be done because switchdown does not occur instantaneously; even when the input intensity I_I exceeds the minimum value $I_\downarrow$ required for switchdown, there is a finite time for the positive feedback to actuate the switching—in the experiment here by heating the medium and shifting the absorption to lower energy. During this time I_I continues to increase, exceeding $I_\downarrow$ farther into the sample. The switchdown of the front part is seen as a sudden decrease in the light reaching the back part and in the light exiting the sample; if the carrier density and temperature of the back part adiabatically follow the light intensity, they too will decrease moving away from their critical values for switchdown. But if the input continues to increase, eventually the next portion of the sample switches down giving rise to a sudden decrease in transmission and indicating that the kink has jumped to the boundary between the second "off" portion and the remaining "on" portion.

Most observations of increasing absorption have been made with samples so short that longitudinal heat conduction and/or carrier diffusion prevented the observation of kinks. To avoid that problem a sharp-cut color filter consisting of semiconductor crystallites embedded in glass is used here. The low concentration of crystallites maintains a relatively sharp band edge while eliminating carrier diffusion and reducing the absorption per unit pathlength by three or four orders of magnitude relative to that for a pure crystal. Millimeter lengths of color-filter material are then required for 10%–50% absorption. Such lengths are much more convenient for filter construction and result in much longer conduction times across the samples making kinks possible.

The observation of kinks is made using a Corning CS3-70/No. 3384 sharp-cut color filter, ground into a wedge and polished. The wedge eliminated cavity effects; translation of the wedge varied the thickness from 50 μm to 3 mm. The 514.5-nm input beam was varied from 0 to 1 W in 20 ms to 4 s and focused to a diameter of 10 to 40 μm. Entire-sample increasing absorption optical bistability was observed with a rise time of 250 ms and a thickness of 200 μm. The bistability involves a thermal shift of the band edge. Figure 1(a) shows the room-temperature band edge $\mathscr{T}(\lambda)$ of a 0.2-mm-thick filter, and the temperature dependence of the 514.5-nm transmission $\mathscr{T}(T-T_0)$ is shown in Fig. 1(b). The straight lines A and B indicate the values of input intensity for switchdown $I_\downarrow$ and switchup $I_\uparrow$ according to the usual steady-state graphical description[4] of increasing absorption bistability.

Kinks are observed in an $\simeq$2-mm-thick filter, as shown in Fig. 2. A single sudden drop in I_T may result from either entire-sample or partial-sample switchdown. But two or more sudden drops assure that a kink has jumped deeper into the sample. The sequence (a) to (c) in Fig. 2 shows fewer kinks the faster the input rise time. Pronounced changes (most noticeably blooming) in the far-field profile occur, but they are suppressed here by collecting all of the transmitted light.

The color-filter kinks are modeled using the transport equations of the intensity I, the temperature T, and the

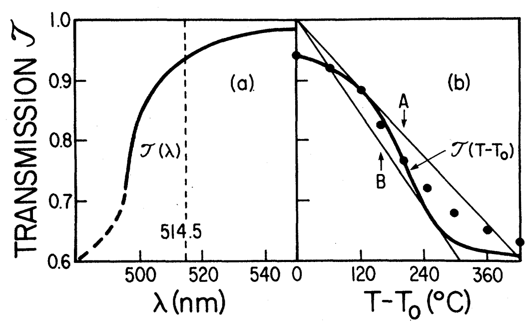

FIG. 1. Transmission $\mathcal{T}$ of a Corning CS3-70/No. 3384 sharp-cut filter with thickness $L \simeq 0.2$ mm as a function of wavelength λ at $T = 23$ °C and as a function of temperature ($T - T_0$ with $T_0 = 22$ °C) for $\lambda = 514.5$ nm. The circles are theoretical values of $\mathcal{T}(T-T_0) = \exp[-\alpha(514.5 \text{ nm}, N_{eh}=0, T=T_0)L]$ from Eq. (5) using $L = 0.25$ μm. The dashed lines A and B correspond to the experimental critical intensities for $I_\downarrow$ and $I_\uparrow$, respectively.

electron-hole density N_{eh} in a medium with an absorption coefficient $\alpha(\omega, N_{eh}, \Delta T)$ and refractive index $n(\omega)$:

$$\frac{\partial I}{\partial t} = -\frac{c}{n}\left(\alpha I + \frac{\partial I}{\partial z}\right) , \tag{1}$$

$$\frac{\partial T}{\partial t} = \frac{N_{eh} \hbar \omega q}{\tilde{C} \tau_{eh}} - \frac{T}{\tau} + D \frac{\partial^2 T}{\partial z^2} , \tag{2}$$

and

$$\frac{\partial N_{eh}}{\partial t} = \frac{\alpha I}{\hbar \omega} - \frac{N_{eh}}{\tau_{eh}} , \tag{3}$$

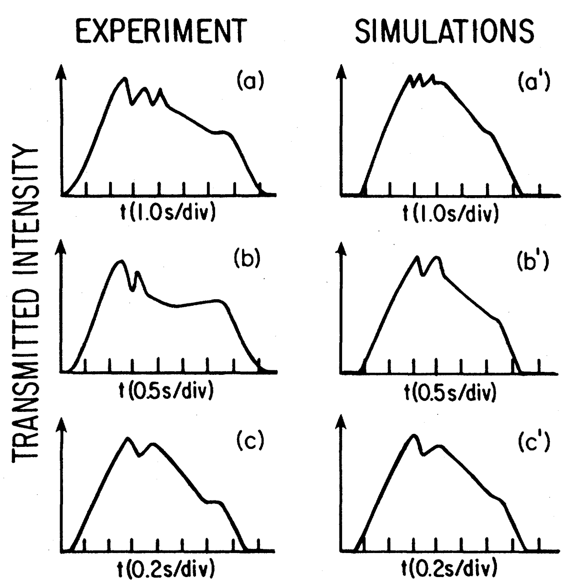

FIG. 2. (a)–(c) Experimental kinks in a Corning CS3-70/No. 3384 sharp-cut color filter of thickness $L \simeq 2$ mm and $r_0 \simeq 14$ μm for the same peak input intensity. The rise time of the triangular input pulse is decreased from (a) to (c). (a')–(c') Numerically calculated normalized $I_I(t)$ and $I_T(t)$ for three different values of $\Delta t/\tau$: (a') 350, (b') 175, and (c') 70. The thickness of $CdZn_xS_{1-x}$ is taken as 2.5 μm suggested by Fig. 1, and the roughly 10 times thicker experimental thickness here.

where $\tilde{C}$ is the product of the specific heat C and the density ρ, τ_{eh} the lifetime of the electrons and holes, q the percentage of nonradiative electron-hole recombinations, $\Delta T = T - T_0$, T_0 room temperature, τ the transverse thermal conduction time, and D the longitudinal thermal conduction coefficient. The carriers are confined to microcrystallites and cannot diffuse. Kinks are most pronounced in the limit of vanishing longitudinal conduction ($D = 0$).

Equations (1)–(3) have been solved numerically using the following model for $\alpha(\omega, N_{eh}, \Delta T)$. The complex dielectric function of the band edge of the $CdZn_xS_{1-x}$ crystallites in the color filters can be written as

$$\epsilon(\omega) = -\epsilon_\infty \frac{e^2 E_g \sqrt{2m}}{\pi} \times \int_0^\infty \frac{\sqrt{y}\,[1 - f_e(e_e) - f_h(e_h)](\omega - E_g' - y - i\gamma)\,dy}{(E_g + y)^2[(\hbar\omega - E_g' - y)^2 + \gamma^2]} , \tag{4}$$

so that

$$\alpha(\omega, N_{eh}, \Delta T) = \frac{\omega}{c n(\omega)} \operatorname{Im} \epsilon(\omega) , \tag{5}$$

where

$$f_i(e_i) = \{1 + \exp[(e_i - \mu_i)/kT]\}^{-1} ,$$

$$E_g' = E_g - \Delta(N_{eh}) - E_T ,$$

$$\Delta(N_{eh}) = E_b[4.78(a_0^3 N_{eh})^{0.25} + 2(a_0^3 N_{eh})^{0.5}] ,$$

$$E_T = (\Delta T) \cdot (0.38 \text{ meV/°C}) .$$

The chemical potentials μ_i are calculated from $N_{eh} = 2\Sigma f_i(e_i)$. In the numerical solutions of Eqs. (1)–(3) longitudinal conduction is neglected ($D = 0$), and transverse conduction enters only through τ. Then intensity $I(z,t)$ adiabatically follows the input intensity $I_I(t)$, so $\partial I(z,t)/\partial t$ is set to zero. Other parameters are $T_0 = 22$ °C, $E_g(T_0) = 2.51$ eV, $q = 0.95$, $m = m_e m_h/(m_e + m_h)$, $m_e = 0.205 m_0$, $m_h = 1.1 m_0$, $\epsilon_\infty = 8$, $\gamma = 10$ meV, $\tau_{eh} = 0.1$ ns, $\tau = 10$ ms,[16] $\hbar\omega = 2.46165$ eV (514.5 nm), and $a_0 = 26$ Å is the exciton Bohr radius. $\tilde{C}$ increases gradually from 1.6 J/(cm^3 °C) at $\Delta T = 0$ to 2.55 at $\Delta T = 500$ °C.[17] The circles in Fig. 1 show the transmission $\mathcal{T} = \exp[-\alpha(\omega, N_{eh}, \Delta T) L_{\text{eff}}]$, where α is calculated from Eqs. (4) and (5). The effective optical absorption length $L_{\text{eff}} = 0.25$ μm of the $CdZn_xS_{1-x}$ microcrystallites in the glass matrix has been obtained by equating $\alpha(\omega, N_{eh}, \Delta T) L_{\text{eff}} = \alpha_{\exp} L$ for a filter with length $L = 0.2$ mm at $T = T_0$.

The temporal variation of the transmitted intensity calculated from Eqs. (1)–(3) is shown in Figs. 2(a')–2(c') for three different rise times Δt of the exciting laser pulse. Both the calculated transmission $\mathcal{T}$ [circles in Fig. 1(b)] and the calculated sawtooth structures of the transmitted intensity (Fig. 2) are in good qualitative agreement with the corresponding experimental findings. The analysis shows, for the present experimental conditions in which no kinks are observed for pulses shorter than 1 ms, that the contribution of the electron-hole pairs to the band-gap renormalization is small compared with the thermal shift.

The experimentally and theoretically found dependence of the time variations of the transmitted intensity on the pulse rise time Δt and on the sample length L can be understood in terms of the following qualitative considerations: The velocity with which the critical temperature $T_{\downarrow}$ for switchdown moves through the filter is roughly $1/(\alpha\Delta t)$. In the time τ a region of length $\Delta L = \tau/(\alpha\Delta t)$ switches to the highly absorbing state. Thus, the number of transmission discontinuities increases and their amplitude decreases if the rise time Δt is increased (Fig. 2). The transverse conduction time τ can be lengthened by defocusing the beam maintaining a constant peak power. It has been checked experimentally that such a procedure indeed yields a reduction of the number of transmission discontinuities. The condition for observation of at least one sawtooth structure is clearly that the kink-jump length ΔL is smaller than the crystal length. This condition has also been checked by doubling the thickness of the filter used for the entire sample bistability: Two sudden decreases in the transmitted intensity were observed indicating partial-sample bistability and jumping of a kink.

Thermal conduction is believed to be responsible for the downward slope in the output signals in Fig. 2(a)–2(c); gradual heating of the entire filter reduces the transmission. In the calculations, switchdown always occurs at the same output intensity, because longitudinal conduction is not included. For $\tau << \Delta t$ and low absorption this means that the temperature of the next section adiabatically follows the output intensity.

In summary, increasing absorption optical bistability has been seen in a Corning CS3-70/No. 3385 sharp-cut color filter. A longitudinal discontinuity (kink) is observed to make sudden jumps deeper into the absorbing sample as the input is increased linearly in a time longer than the transverse conduction time. Observed features of the kinks are in qualitative agreement with numerical calculations based upon a microscopic expression for the band-edge absorption. Kinks are a manifestation of the local character of increasing absorption bistability in that not all of the sample switches down at the same time; therefore, they do not exist for bistability relying upon a cavity or external feedback.

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[16]For a given rise time Δt and peak intensity, the number of kinks is determined by the ratio of $\Delta t/\tau$. If a laser beam suddenly heats a cylinder of length much greater than the beam radius r_0, a solution of the heat diffusion equation leads to $\tau = C\rho r_0^2/[(2.40)^2 K]$, where C is the specific heat, ρ the mass density, and K the thermal conductivity. With $C = 1.02$ J/(g °C), $\rho = 2.5$ g/cm^3, $r_0 = 14$ μm, and $K = 10.04\times 10^{-3}$ W/(cm °C), one has $\tau = 85$ μs. The value $\tau = 10$ ms was chosen to give the same number of kinks as observed in Fig. 2; it corresponds to $r_0 = 150$ μm. This value appears plausible since the steady-state radius of the temperature distribution is clearly larger than the laser beam radius.

[17]Heraeus Quarzschmelze brochure Q-A 1/112.2, 1983, p. 14.