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Nonlinear Absorbers of Light

A nonlinear absorber of light is a substance having an unique property: its optical absorptivity is altered by light irradiation. The phenomenon is known as *photochromism* when the changes in optical absorbtivity are in such a wavelength range and on such a time scale as to be visually apparent. Photochromism has long been a plague to dye manufacturers. It has recently been the basis of many curious and ingenious inventions.[1] The present Communication is inspired by the possible use of nonlinear absorbers in various sorts of data processing applications, including multilevel logic devices. Some quantitative results pertaining to such systems are developed for a model nonlinear optical absorber and presented here.

The model nonlinear absorber to be considered consists of a solid containing a density N of centers which have the energy level structure shown in Fig. 1. A center in the ground state A has a cross section σ for the absorption of a photon of energy $h\nu$. The absorption process raises the electron to level B, from which it immediately falls to state C. In state C it has no appreciable absorption probability for photons of energy $h\nu$ and makes no contribution to the optical density of the material. The electrons in state C decay spontaneously with a time constant τ to state A. In this system, high-intensity light is transmitted efficiently because most of the centers are maintained in the excited state C in the presence of intense radiation. It is useful only when such a property is desired, e.g., in data processing applications, where high absorption of intense radiation creates large amounts of heat. Nonlinearity of the type which transmits the more intense radiation and absorbs the weaker radiation is thus more desirable than the opposite type, in which low-intensity radiation is transmitted and high-intensity radiation is absorbed. Model systems of the latter type, however, can also be constructed, as will be briefly mentioned later.

Referring to Fig. 1, let n be the density of centers in the excited state C and let $(N - n)$ be the density in the ground state A. There are no centers in state B because the decay from state B to state C has been assumed to be instantaneous. Let a beam of light of intensity I photons/cm^2 sec be passing through the absorber and let x measure distance in the absorber in the direction of the beam. Then the change in intensity of the beam with distance is

$$dI/dx = -I(N - n)\sigma. \tag{1}$$

The time rate of change of n is

$$dn/dt = I(N - n)\sigma - n/\tau \tag{2}$$

and dn/dt vanishes in the steady state. Thus

$$n = \frac{N}{1 + (I\sigma\tau)^{-1}}. \tag{3}$$

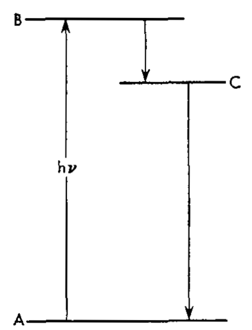
Another system which is phenomenologically identical to that shown in Fig. 1 (i.e., described by the same set of rate equations) could be constructed without the state C. The electrons would then be allowed to decay from state B back to state A with time constant τ. The optical density would be reduced by the accumulation of electrons in state B because the interaction of photons with the electrons in state B would increase the intensity of the photon beam by stimulating the decay to state A and the accompanying emission. The equations for this model are identical to Eqs. (1) and (2) if N is replaced by $(N/2)$ and σ is replaced by 2σ in Eqs. (1) and (2).

The quantity $(\sigma\tau)^{-1}$ is essentially the unit of intensity throughout the problem and is designated here by

$$J = (\sigma\tau)^{-1}. \tag{4}$$

Note that $J = \infty$ characterizes a linear absorber. Thus

Figure 1 **Energy levels of a model nonlinear light absorber.**



Eq. (3) can be written $n = N/(1 + J/I)$.

It is already possible to recognize an important feature of the results. If I is much smaller than J, then n is much smaller than N. Eq. (1) then becomes $dI/dx = -IN\sigma$, the description of a linear absorber. Nonlinear effects occur only when I is comparable to or larger than J, and the required intensity obviously depends on τ, the response time of the system. The value of J increases with decreasing τ.

For example, take 10^{-17} cm^2 as an attainable value for σ, the absorption cross section. Values of τ of 10 sec or longer would be satisfactory for many applications involving visual photochromism, such as variable transparency window panes or sun glasses.[1] Then $J = 10^{16}$ photons/cm^2 sec. Suitable devices for this class of application can be constructed, since sunlight contains over 10^{17} photons/cm^2 sec.

On the other hand, values of τ of 10^{-9} sec or less are desirable for high-speed data processing applications. Then $J = 10^{26}$ photons/cm^2 sec. This magnitude of photon density is not available from any light source other than the giant pulse of a ruby laser. It appears that the use of nonlinear absorbers in high-speed data processing will not be practical unless systems with $\sigma > 10^{-13}$ cm^2 can be found.

Nevertheless, it is worth while to pursue the solution of Eqs. (1) and (3) further. There are possible intermediate-speed applications to memory, display, slow data processing, and protection against radiation.[1] Substituting Eq. (3) into Eq. (1) gives the differential equation $(\sigma\tau + I^{-1})(dI/dx) = -N\sigma$. The solution is

$$\log (I/I_0) + (I_0/J)[(I/I_0) - 1] = -N\sigma x. \tag{5}$$

Here I_0 is the value of I at $x = 0$ and J has been introduced from Eq. (4). $x = 0$ will be regarded as the boundary of the absorber from which the light intensity I_0 enters. A plot of I/I_0 as a function of distance into the absorber is given in Fig. 2 for several values of the parameter I_0/J.

It is seen from Eq. (5) that a nonlinear absorber is described by two parameters. One is J, which is a constant of the material since σ and τ are constants of the system. The other is the low-level optical density, $N\sigma x$, which can be controlled by varying N, the concentration of centers, or x, the thickness, or both. The problem of design of a nonlinear absorber is that of selecting a value of $N\sigma x$ which gives sufficient difference between the transmission of high- and low-level incident light and holds the absorption of the high-level light, which it is desired to transmit, to an acceptable level. It will be seen that these are conflicting requirements. The way in which the compromise between them will be made depends on the particular application in question and on the range of intensities of the incident signals.

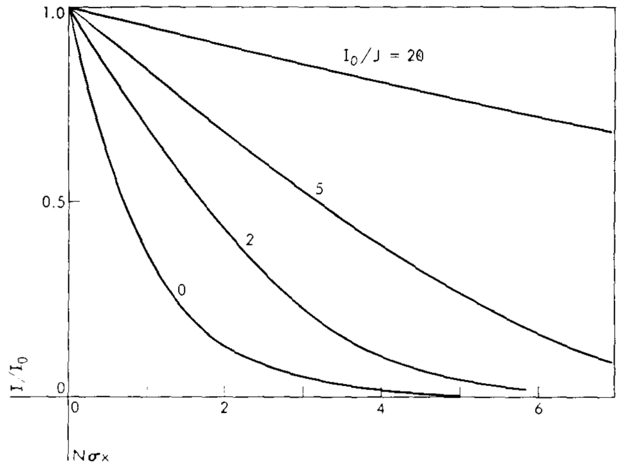


Figure 2 **The form of the absorption curves defined by Eq. (5).**

It is not feasible to present a general analytical exhibition of this point. A couple of examples will illustrate it, however.

First, consider that either a strong beam of intensity $I_0'' \gg J$ or a weak beam of intensity $I_0' \ll J$ may be incident on the absorber. Let the emergent beams have intensities, respectively, of I'' and I'. Eq. (5) reduces to

$$I'' = I_0'' - N\sigma x\, J \tag{6}$$

for the strong beam and to

$$I' = I_0 \exp(-N\sigma x) \tag{7}$$

for the weak beam. The attenuation of both beams increases with increasing optical density, $N\sigma x$. The desired attenuation of the weak beam, Eq. (7) can be obtained only if some absorption of the strong beam is also present. The relation between the attenuation of the weak beam and that of the strong beam can be expressed quantitatively by eliminating $N\sigma x$ from Eqs. (6) and (7): $I' = I_0' \exp [-(I_0'' - I'')/J]$. Strong attenuation of the weaker beam requires that the loss of intensity of the stronger beam be several times J.

Second, consider the small-signal rate of change of (I/I_0) with I_0. This is conveniently characterized by the power with which I depends on I_0, i.e., $d(\log I)/d(\log I_0)$, which we designate by G, a figure of merit for nonlinearity. It is equal to unity for a linear absorber. Differentiating Eq. (5) with respect to I_0 gives $G = (J + I_0)/(J + I)$, which illustrates both of the points made above. It is necessary that I_0 be large compared to J if G is to be large. It is also necessary that I be small compared to I_0.

Photochromism of other types can be produced in the present model by attributing different properties to an electron in the excited state C. For example if, instead of assuming that an excited center is optically inactive, it had been assumed that a center in state C had a much larger cross section for absorption than a center in state A, then high-intensity light would be more strongly absorbed than low-intensity light. The nonlinearity would be of the opposite type. Or the absorption by a center in state C would be at a different energy than that of state A. Then irradiation at one frequency would alter the absorption at a second frequency.

In summary, two important features emerge from the theory of the model nonlinear optical absorber: (1) Nonlinear effects occur only if the incident light intensity is large compared to $(\sigma\tau)^{-1}$, where σ is an optical absorption cross section and τ is a relaxation time which determines the response time of the system. (2) There must be appreciable absorption of the beam which is intended to be transmitted efficiently.

Reference

1. G. Dorion and I. Weissbein, *Discovery* 24, No. 2, 32 (1963).

Received May 20, 1963