Intrinsic Optical Bistability in Nonlinear Media

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A general class of optically bistable systems is described which operates without feedback and is based on the intrinsic response of certain nonlinear media. A detailed analysis of one such medium is presented which demonstrates that bistability may result in virtually all macroscopic optical and material parameters.

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Within the past decade optical bistability has grown into one of the most active research areas in nonlinear optics.[1] The standard method for obtaining a bistable optical device requires a nonlinear optical interaction coupled with an intrinsic (optical) or hybrid (electrical) feedback mechanism. In such systems the nonlinear medium responds uniquely to the impressed optical (and static) fields. Bistability results from a multiplicity of values for the total impressed field due to a nonunique feedback which is usually proportional to the output field. Thus, the system response is not uniquely specified in terms of the input alone. Recently several systems have been proposed[2-4] and demonstrated[5-8] which we believe to be examples of a large class of bistable devices which operate on an entirely different basis. In this Letter we describe the general mechanism by which these systems operate and also provide a detailed analysis of the macroscopic effects due to one particularly simple model for such media. Since these media may also display numerous other nonlinear processes such as harmonic generation, wave mixing, and self-focusing effects, when intrinsic microscopic bistability occurs a reformulation of the standard methods of analysis for these other nonlinear processes is required.

When a nonlinear medium is excited by an optical field, the microscopic response of the medium may be represented through one or more nonlinear constitutive relations of the form

$$f(\vec{E},\vec{a}_i,b_i)=0, \qquad (1)$$

where $\vec{E}$ represents the impressed fields and $\vec{a}_i$ and b_i are vector and scalar microscopic response parameters. The standard procedure in handling relations of this form is to expand the macroscopic parameters determined by $\vec{a}_i$, and b_i in power series in the fields, such as is done in the polarization expansion. These macroscopic parameters are then used in Maxwell's equations to produce nonlinear wave equations, whose solutions must be consistent with Eqs. (1) and the relevant boundary conditions.[9] This procedure assumes that the optical fields may be treated as independent variables. However, under appropriate conditions this assumption breaks down and relations (1) lead to a multivalued response wherein two or more stable solutions for $\vec{a}_i$ and/or b_i may exist over a range of values for $\vec{E}$. In such cases the nonlinear wave equation must be solved in terms of the material parameters as the independent variables, which are then related to the optical fields through Eqs. (1).

Specific examples of the multivalued nonlinear constitutive relations (1) have recently been demonstrated for absorptive bistability due to band-gap renormalization[5] and to thermal variation of the band gap in semiconductors,[6,7,10] and in the state of liquid crystals.[8] Proposals for bistability due to relativistic mass effects in the cyclotron resonance of electrons,[2] to the local-field correction[3] and to atomic correlation in small volumes[4] have also appeared. The essential (and somewhat unusual) notion that the driving fields must be treated as dependent on the material response was first realized in a related context by Duffing in his work on the classical anharmonic oscillator.[11] In order to explicate the general idea and to indicate the range of macroscopic manifestation of intrinsic material bistability on the optical driving fields we consider a simple classical model for the complex nonlinear polarization based on the Duffing oscillator which has been previously employed in the analysis of bistability in four-wave mixing.[12] Here we describe both absorptive and dispersive optical bistability with only a single propagating wave.

Consider plane-wave propagation in the $+z$ direction from a linear medium $(z<0)$ with index n_L into a nonlinear medium $(z\geq 0)$. For mathematical convenience we restrict the discussion to low molecular densities N, and the polarization of the nonlinear medium at the fundamental frequency ω is taken to be $\vec{P}(z,t)=\vec{P}(z)e^{-i\omega t}=N\vec{p}(z,t)\equiv N\vec{p}(z)e^{-i\omega t}$ where $\vec{p}(z)$ is the amplitude of molecular polarization and the driving field is $\vec{\mathscr{E}}(z,t)=\vec{\mathscr{E}}(z)e^{-i\omega t}$. The differential form of the constitutive relation is taken to be

$$\frac{m}{e}\frac{\partial^2\vec{p}}{\partial t^2}+\gamma'\frac{\partial\vec{p}}{\partial t}=-\vec{\nabla}V(\vec{x})+e\vec{E}(z,t), \qquad (2)$$

where m is the mass of the bound charges and $\gamma\partial\vec{p}/\partial t$ represents a linear loss. The one-dimensional Duffing potential $V_D=\frac{1}{2}\kappa x^2+\frac{1}{4}\beta' x^4$ is the simplest binding potential which leads to bistable response. Substituting $V_D(x)$ in Eq. (2) leads directly to the solution to order β at the fundamental frequency,

$$[(\Delta-i\gamma\omega)+\tfrac{3}{4}\beta|p|^2]p=\frac{e^2}{m}\mathscr{E}, \qquad (3)$$

and an additional relation for the third harmonic, which we ignore. Here $\Delta\equiv\omega_0^2-\omega^2$, $\omega_0^2\equiv\kappa/m$, $\gamma\equiv\gamma'/m$, and $\beta\equiv\beta'/me^2$. Figure 1(a) shows the bistable response of $|\vec{P}|$ versus average flux $\bar{S}$ based on Eq. (3).

The essential point in our analysis now lies in treating $\vec{p}$ (or equivalently, $\vec{P}$) as the independent parameter in the wave equation. With Eq. (3) defining $\mathscr{E}=\mathscr{E}(z)$, the wave equation may now be solved for $p(z)\equiv\Lambda(z)\exp\{i[\phi(z)+kz]\}$. The slowly varying envelope solution is easily obtained:

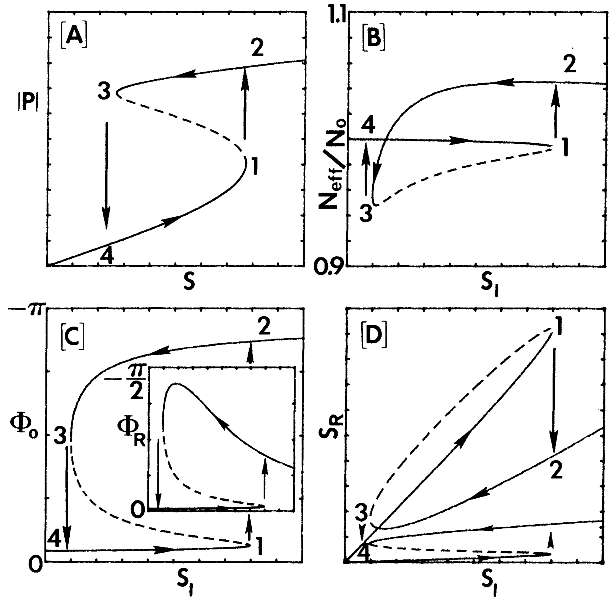

FIG. 1. (a) Polarization amplitude vs flux $\bar{S}$ for a medium modeled by Eq. (2) with a Duffing potential V_D, which leads to (b) a bistable effective index of refraction n_{eff}, (c) a bistable phase relation Φ_0 (Φ_R, inset) between the incident and transmitted (reflected) field, and (d) a bistable reflected flux $\bar{S}_R$ obtained through interference with a reference beam, all vs incident flux $\bar{S}_I$. Conditions for bistable behavior are: (1) $(\omega_0^2-\omega^2)/\beta<0$, and (2) $(\omega_0^2-\omega^2)^2>3\gamma^2\omega^2$, where ω_0 is the microscopic resonance frequency, ω is the field frequency, $\beta=\beta'/me^2$ is the coefficient of the quartic potential term, and $\gamma=\gamma'/m$ is the damping coefficient in Eq. (2). The switchup intensity [$1\rightarrow 2$ in (a)] is $\bar{S}_{sw}=-2m^2c(\omega_0^2-\omega^2)^3/81\pi e^2\beta$.

$$\ln[\Gamma(z)/\Gamma_0]+\{\tfrac{3}{2}\beta\Delta[\Gamma^2(z)-\Gamma_0^2]+\tfrac{27}{64}\beta^2[\Gamma^4(z)-\Gamma_0^4]\}[\Delta^2+\gamma^2\omega^2]^{-1}=-\alpha z, \qquad (4)$$

and

$$\phi(z)=\phi_0+(1/2k)[\omega^2/c^2-k^2]z-(1/\gamma\omega)\{\Delta\ln[\Gamma(z)/\Gamma_0]+\tfrac{9}{8}\beta[\Gamma^2(z)-\Gamma_0^2]\}, \qquad (5)$$

where, for small loss, k and α satsify the usual (linear oscillator) conditions:

$$k=\frac{\omega}{c}\left[1+\frac{\omega_p^2\Delta}{\Delta^2+\gamma^2\omega^2}\right]^{1/2}\equiv\frac{n_0\omega}{c}, \qquad (6)$$

and

$$\alpha=\gamma\omega^3\omega_p^2[2kc^2(\Delta^2+\gamma^2\omega^2)]. \qquad (7)$$

Here Γ_0 and ϕ_0 are the amplitude and phase of $p(z)$ at $z=0$ and $\omega_p[4\pi Ne^2/m]^{1/2}$ is the plasma frequency of the material.

Equations (4) and (5) determine $p(z)$ and Eq. (3) then yields $\mathscr{E}(z)$ from which the energy flux in the nonlinear medium is obtained:

$$\bar{S}=(cm^2\Gamma^2/8\pi e^2)\{[n_0+(c\phi'/\omega)][(\Delta+\tfrac{3}{4}\beta\Gamma^2)^2+\gamma^2\omega^2]+\tfrac{3}{2}\beta\gamma c\Gamma\Gamma'\}, \qquad (8)$$

where $\Gamma'\equiv d\Gamma/dz$ and $\phi'=d\phi/dz$ are obtained from differentiation of Eqs. (4) and (5). The incident flux $\bar{S}_I$ may now be related to $P(z=0)$ through standard boundary conditions at $z=0$ by utilizing Eq. (3) for

$\mathscr{E}(z=0)$. For normal incidence we obtain

$$\bar{S}_I=\left(\frac{cn_L}{8\pi}\right)\left(\frac{m^2\Gamma_0^2}{16n_0^2n_L^2e^2}\right)\{[\gamma\omega(n_0^2=2n_Ln_0+1)]^2+[(n_0^2+2n_L+1)(\Delta+\tfrac{3}{4}\beta_0^2)+\omega_p^2]^2\}. \tag{9}$$

The reflected flux $\bar{S}_R$ is obtained in a similar fashion.

The relations between incident, transmitted, and reflected flux are obtained by varying Γ_0 simultaneously in $\bar{S}_I$, $\bar{S}$, and $\bar{S}_R$. An effective index may be defined in terms of the effective wavelength, or by the relation between $\mathscr{E}$ and P. Figure 1(b) shows the effective index n_{eff} at $z=0$ versus incident flux. The transmitted flux at $z=0$ is then determined (for small loss) by $\bar{S}=[4n_{\text{eff}}n_L/(n_L+n_{\text{eff}})^2]\bar{S}_I$. The phase difference between the incident field and the polarization at $z=0$ must also be expressed in terms of Γ_0:

$$\tan\phi_0=\frac{(2n_0n_L+n_0^2+1)\gamma\omega}{(2n_0n_L+n_0^2+1)(\Delta+\tfrac{3}{4}\beta\Gamma_0^2)+\omega_p^2}. \tag{10}$$

Figure 1(c) shows this phase difference versus incident flux again obtained by varying Γ_0 simultaneously in ϕ_0 and $\bar{S}_I$. The large phase jumps at switching are characteristic of this type of polarization bistability. The inset in Fig. 1(c) shows the reflected-field phase shift versus incident flux. As expected, the reflected-field phase jump at switching is large only when n_{eff} crosses (or approached) n_L.

As the transmitted field propagates in the nonlinear medium the field amplitude decays at different rates for the two stable branches according to Eq. (4) (bistable absorptivity). An abrupt change in the effective index $n_{\text{eff}}(z)$ within the material will occur if, at some z, the intensity drops below the minimum value needed to maintain the upper branch polarization leading to self-reflected waves. Similar effects should also occur in nonplanar wave propagation, such as self-enhanced, self-focusing, and self-defocusing. The total phase shift of the optical field over a length of the nonlinear medium is determined by Eqs. (4) and (5) and also displays two stable values, as does the transmitted field at the nonlinear to linear output interface. The net effect of the amplitude and phase bistability at the input and output interfaces plus the bistable phase shift and absorption across the medium itself leads to a net bistability in both the amplitude and phase of the transmitted field across a slab of the nonlinear material. For physically reasonable nonlinearities the amplitude variations should be quite small, but the phase variations are not. The phase bistability may be converted into a flux bistability through any standard interference configuration. As pointed out above, phase bistability upon reflection is substantial when n_{eff} varies in the neighborhood of n_L, and thus may also produce flux bistability through interference with a reference beam. This effect is shown in Fig. 1(d) for two different low-intensity phase differences between the interfering beams. The bistable loops displayed in Figs. 1(b) and 1(d), which may also occur in other macroscopic parameters, result from the fact that each macroscopic parameter has a multivalued response with the respect to the independent material variable. However, the fundamental material response curve [Fig. 1(a)] always maintains its characteristic "S shape".

It must be stressed that the standard procedures of nonlinear optics which result in power series expansions in terms of the electric fields cannot be used to describe this type of bistability. The potentially important class of bistable interactions which result from intrinsic material bistability must be treated by procedures similar to the one outlined above. Then the problem is reduced to the discovery of appropriately nonlinear constitutive relations.

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