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MICOM PROGRAM IN OPTICAL BISTABILITY (U) CHARLES M. BOWDEN AND ADVID W. HOWGATE Dr.* US ARMY MISSILE COMMAND REDSTONE ARSENAL, ALABAMA 35809 I. INTRODUCTION

Some years ago, a number of people (1-4) independently suggested the use of a saturable absorber to induce optical bistability (hereafter referred to as OB). The phenomenon was demonstrated and studied recently by Gibbs, McCall and Venkatesan (5) who used a cw dye laser to excite atoms of sodium vapor in a cell between the plates of a Fabry Perot interferometer. Under certain experimental and material conditions, they observed a nonlinear dependency of the transmitted field as a function of the dye-laser input which exhibited hysteresis, differential gain, and bistable behavior.

Interest in OB from the practical applications standpoint stems from obvious device applications as the optical analog of the transistor, optical clipper or limiter and digital optical memory element. The recent demonstration of the phenomenon (5) has generated considerable interest from the fundamental point of view as a clear example of spontaneous ordering in an open, stationary system of matter interacting with light. This has led to a surge of recent theoretical activity.

The first model for OB was proposed by McCall (6), who introduced a nonlinear susceptibility into the Maxwell-Bloch representation in the full propagation treatment. His results are necessarily calculational rather than analytical. Bonifacio and Lugiato (7) were the first to obtain the main features of OB from implementation of the Maxwell-Bloch model, which they solved analytically in closed form. They later extended their model (8) to a quantum-statistical representation of a stationary system far removed from thermodynamic equilibrium, but with the same essential assumptions, the most crucial being

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the "mean field" approximation which amounts to requiring that the field be sufficiently uniform over the volume of active atoms. Others, such as Walls et al. (9) have considered similar models which are, on the whole, in agreement with the results of Bonifacio and Lugiato.

All of these models have one essential common characteristic, namely, they consider OB within the framework of individual or independent atom response to the electromagnetic field. These are therefore necessarily absorptive models which describe OB as a transition from a state of coherent response of individual atoms to the externally-applied driving field and high absorption (low transmission) to a state of saturation or bleaching of the material (high transmission). The experimental results on sodium vapor (5) exhibited a large dispersive contribution, and these models do not appear to describe the dispersive aspects even qualitatively, although the absorptive characteristics appear to be adequately qualitatively described.

It seemed to us that OB should stem, at least under certain conditions, from the same kind of fundamental interaction that gives rise to the dynamical cooperative process known as superfluorescence (10, 11). This prompted us to take a fresh approach to the interpretation of OB in terms of a model which emphasizes the contribution from interatomic cooperative processes in matter interacting with light. Our model (12) differs from other previous models in that it includes the possibility for atom-atom interaction and correlation via the mutual internal electromagnetic field (virtual photon exchange) as well as the individual atom coupling to the electromagnetic field.

The results of our model (12) and other models (7-9) predict identical behavior for "absorptive" OB (perfect cavity, laser and atomic tuning). However, our results fit the dispersive characteristics reported for the sodium experiment (5) qualitatively quite well. In addition, we predict new results in reversible hysteresis and bistability in the variation of inverse effective temperature T_s with the output field for fixed input above a certain threshold value. This new result (13) has a wide range of potential applications, including a sensitive IR, FIR or millimeter wavelength detector with frequency upconversion.

In addition, our results provide an interpretation of OB as a true first-order phase transition, from atomic order to disorder due to virtual photon exchange between atoms, far from thermodynamic equilibrium. In the absence of an applied field, our results show the existence of a second-order phase transition (12,14) below a critical temperature determined by material parameters and cavity detuning.

Another unique characteristic of our results is that the model constitutes the optical analog of the Meisner effect (15) in superconductivity. Due to the collective atomic behavior (optical Meisner effect), our results predict for the first time the existence of OB in a small volume without mirrors. This could have far-reaching applications in the use of arrays of micron size elements for optical digital information storage and retrieval as well as optical digital image formation.

Finally, it should be pointed out that this work is a direct outgrowth of our program in swept-gain superradiance (16-18). Furthermore, MICOM will co-host, with ARO-D, an International Conference and Workshop on Optical Bistability, in Asheville, North Carolina, 3-5 June 1980. This conference is expected to significantly advance the field and to provide a timely record of the state-ofthe-art in this rapidly-developing and potentially extremely useful field. The conference and its impact are anticipated to be similar and of much the same significance as the conference hosted by MICOM on Superradiance (19).

II. THEORETICAL MODEL AND INTERPRETATION OF PHENOMENON

We consider a collection of N identical two-level atoms with energy level separation ε interacting with the electromagnetic field in the dipole approximation. The electromagnetic field which interacts with the atoms is comprised of an internal field and, in addition, an applied cw field assumed to be in a coherent state (20), with field amplitude α and carrier frequency ω_0 . The atoms of the material are considered to be confined to a volume Vg which is much smaller than the resonance wavelength λ_r associated with the atomic bare-state separation. This assumption does not critically affect our results and tends to allow emphasis upon the nature of collective atomic aspects in the results in an unencumbered fashion.

The Hamiltonian H which represents this model is written in the form

$$H = H_{a} + H^{*}$$
(II-1)

where, in units such that $\bar{n} = c = 1$,



$$H' = \left(\sum_{j=1}^{N}\sum_{k=0}^{\infty} g_{k}a_{k}^{\dagger}\bar{\sigma}_{j} + h \cdot c\right) + \left(\alpha \star e^{i\omega_{o}t}\sum_{j=1}^{N}\sigma_{j}^{-} + h \cdot c\right), \quad (II-2b)$$

where $h \cdot c$ means hermetian conjugate, and we have used the rotating wave approximation.

The first and second terms in (II-2a) represent the free field and atomic system, respectively, whereas the first two terms of (II-2b) describe the interaction of the atoms with the internal field and the last two terms describe their interaction with the externally-applied coherent field. The factor α is the complex externally-applied field amplitude. The field photon creation and annihilation operators conform to the Bose commutation relations (12), and the atomic operators are the usual Pauli operators, and

$$g_{k} = -i(\omega_{k}/2V)^{\frac{1}{2}} \underline{\varepsilon}_{k} \cdot \underline{d}$$
 (II-3)

where V is the quantization volume and $\underline{\epsilon}_k$ and \underline{d} are the polarization vector and the matrix element for the transition dipole moment for the atoms (21).

We are interested in determining the steady state properties of the system described by (II-2a, II-2b). In order to retain the essential many-body aspects of the system, it is convenient to treat the system in an ensemble representation. This means taking ensemble averages of relevant dynamical operators such as population difference and dipole moment operators. To facilitate this, we perform the following operations: (i) Canonically transform to a perpendicularly doubly rotating frame, rotating at the laser frequency ω_0 , so the Hamiltonian (II-2) becomes explicitly time-independent (12). The electromagnetic field then appears as a d.c. field in the transformed frame. (ii) Introduce an effective temperature T_g in the transformed representation which is the exact analog of a "spin temperature" (12,22). This is equivalent to a definite statement of the form of the density operator ρ , in the transformed representation.

Although the system is an open system far removed from thermodynamic equilibrium, we can analyze the macroscopic properties of the system in the transformed representation in terms of the free energy F, given by -0 F -0 H

$$e^{-\beta} F -\beta H_{s} F + \beta H_{s$$

where $\beta_{k} = 1/kT_{k}$ and k is Boltzmann's constant.

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Making use of the transformation to the doubly rotating frame (2-4) of reference (12) and adiabatically eliminating the internal field variables, the transformed Hamiltonian H_T , through second order in the atom-field interaction, was shown by Bowden and Sung (12) to be

$$H_{\rm T} = H_{\rm oT} + H_{\rm T}^* \qquad (11-5)$$

where

$$H_{oT} = \Omega a^{\dagger}a + {}^{j}\Omega_{a} \sum_{j=1}^{N} \sigma_{j}^{z} , \qquad (II-6a)$$

$$H_{T}^{*} = -\overline{g}/N \sum_{\substack{j,l=1\\ j \neq l}}^{N} \sigma_{j}^{+} \sigma_{l}^{-} + \alpha \sum_{j=1}^{N} \sigma_{j}^{-} + \alpha^{*} \sum_{j=1}^{N} \sigma_{j}^{+} . \quad (II-6b)$$

Here,

$$\Omega = \omega - \omega_0$$
, $\Omega_a = \varepsilon - \omega_0$. (II-7)

For simplicity, we have considered only a single radiation field mode of frequency ω interacting with the atomic transition. If the atomic medium is in a cavity, then ω is the cavity mode frequency. If there is no cavity, then $\omega = \varepsilon$. The first term in (II-6b) is interpreted as atomic interaction via the electromagnetic field, where the effective atom-atom coupling, \overline{g} , is field-dependent (12),

$$\bar{g} = g_0^2 \frac{(\omega - \omega_0 - \lambda)}{\left[(\omega - \omega_0 - \lambda)^2 + \gamma^2\right]}$$
(11-8)

where

$$g_{0}^{2} = \frac{(4\pi)^{2} d^{2}}{\omega} k \bar{p}$$
 (II-9)

In the above, λ is the Stark shifted atomic level separation (12),

$$\lambda = + \left[n_a^2 + |\Delta|^2 \right]$$
 (II-10)

where $|\Delta|$ is the Stark shift which is dependent upon the value for the internal field intensity. Since the latter is to be determined in the result, $|\Delta|$ must be determined self-consistently later on in the results of the calculation. It is crucial to retain the Stark shift in (II-10) (usually neglected in second-order calculations) in order to obtain the first-order phase transition which leads to OB. The damping term γ appearing in (II-8) is just the cavity width (12) or, if no cavity is present, it is just the photon escape rate from the atomic volume. In (II-9), d is the matrix element of the

transition dipole moment, k is the density of field modes in the neighborhood of the principal mode of frequency ω , and $\bar{\rho}$ is the total number of atoms N in a cubic wavelength at atomic resonance λ_r .

The equation (II-6) is our working Hamiltonian. The atomradiation field interaction (II-2b) has now been recast in terms of an effective interatomic pair interaction via the internal field (II-6b). The pair correlation is depicted by the first term in (II-6b) and may lead to cooperative atomic effects by virtual photon exchange. This term is identical to the interaction which leads to cooperative atomic behavior in superfluorescence, (10,23). The inclusion of the Stark shift correction in \bar{g} , (II-8), (II-10) gives negligible contributions to the results for superfluorescence, but is absolutely essential to arrive at OB in the steady state for this model. This causes the effective interatomic coupling to be internal field intensity dependent, and thus leads to feedback between the atoms and their mutual radiation field. The atomic pair correlation term in the interaction is also identical to that for Cooper pairs in the BCS theory of superconductivity (12,15).

We proceed now to calculate the free energy F from (II-4). To perform the trace on the right-hand side of (II-4), it is necessary to linearize the Hamiltonian (II-6) and to obtain a thermodynamically equivalent linearized Hamiltonian (25,26). The method of references (25) and (26) is used to obtain

$$H_{T}^{*} = N \bar{g} |v|^{2} - N \Delta^{*} \sigma^{*} - N \Delta \sigma^{-}, \qquad (II-11)$$

where

$$\Delta \equiv \overline{g} v^{*} - \alpha^{*} . \qquad (II-12)$$

Here, v is a variational parameter used in the linearization, to be determined by minimization of the free energy F , (25,26).

In the above expressions, we have used the assumption that all the atoms are identical to drop the subscripts, j, and to replace the sums by N and the double sums by N(N-1).

The thermodynamically equivalent Hamiltonian (II-11), i.e., the linearized Hamiltonian which yields the same free energy F in the limit of large N as the original Hamiltonian (II-6b) has an immediate physical interpretation. The interaction expressed in (II-6b) is thermodynamically equivalent to a single atom, mean field interaction expressed in (II-11). The factor Δ , (II-12), is the mean field, i.e., the total field acting on a single atom due to the combined atomic reaction fields of all the other atoms (first term in (II-12) and the local value of the externally-applied field (second term in (II-12)). The expression for the total local field at an atomic site,(II-12),

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is just Maxwell's equation (all electromagnetic fields appear as d.c. fields in the transformed frame rotating at the laser frequency ω_0).

The free energy F is now easily evaluated from (II-4) using (II-11) to give,

$$F = N \tilde{g} |v|^2 - \frac{N}{\beta_s} \log \left[2 \cosh \frac{\beta_s}{2} \lambda\right] + \frac{1}{\beta_s} \log \left(1 - e^{-\beta_s \Omega}\right) . (II-13)$$

The variational parameter v is evaluated from the normal equation, (25,26),

$$\partial F/\partial v = 0$$
 (II-14)

If we let

 $x = \Delta^{\pi}$, $y = -\alpha$ (II-15)

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equation (II-14) yields

$$\frac{\partial F}{\partial v} = N g v^* - 2 \bar{g} x^* \frac{\tanh \frac{s}{2} \lambda}{\lambda} = 0. \quad (II-16)$$

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From (II-16),

$$v = 2 \times \frac{\tanh \frac{\lambda}{2} \lambda}{\lambda}$$
, (II-17)

which is easily shown to be the ensemble average for the macroscopic atomic polarization (12). It is furthermore easily shown that the above condition obtained from (II-14) corresponds to an absolute minimum, i.e.,

$$\partial^2 F/\partial v^2 > 0$$
. (II-18)

It is to be noted that the Stark shift Δ is now determined selfconsistently from (II-12) and (II-17).

If we use the definitions (II-15) in (II-12) and use (II-12) to eliminate v from (II-17), then after multiplying both sides of (II-17) by \bar{g} , the following equation of state for the applied field y as a function of the internal field x is obtained,

$$x - y = 2 \frac{1}{g} x - \frac{t \sinh \frac{p_{\text{II}}}{2} \lambda}{\lambda} . \qquad (\text{II}-19)$$

This is the main result of this paper and leads to bistability, reversible hysteresis and differential gain between the three quantities, x, y, and β . These characteristics will be discussed in the next section.

III. INTERPRETATION OF RESULTS

A. Optical Bistability as a First-order Phase Transition

The main result of our model, (II-19), has the form typical of a first-order phase transition. For absolute zero of effective temperature and for perfect laser and cavity tuning, i.e., for $T_g \rightarrow 0$, $\omega = \varepsilon = \omega_0$, (II-19) reduces to exactly the form for the equation of state obtained earlier from the single-atom, coupled Maxwell-Bloch models (8), (9), i.e.,

$$y = x + 2c \frac{x}{(1 + x)^2}$$
, (III-1)

where

$$c = g_0^2 / \gamma^2$$
, (III-2)

and we have written x and y in units of γ . Here, however, the physics is quite different as will be discussed later.

Equation (II-19) is shown plotted in Figure 1, input field amplitude y vs. output field amplitude x for various values of equal cavity and laser offtuning. It is readily established that the minimal condition for OB to occur is that c > 4, (8,12).

It is easily shown (27) that all points on the curves in Figure 1 are stable steady-state conditions except those for which dx/dy < 0. Thus, the points on the curves between the turning points having negative slope are unstable. Increasing the applied field y from zero, the output follows a nearly linear response until the first turning point is reached along the curve for the equation of state. In this regime, the material responds mainly by atom-atom coupling via virtual photon exchange, i.e., \overline{g} , (II-8) is large due to the relatively small Stark shift (II-10). The reaction field due to the collective response of the atoms (first term on the right-hand side of (II-12)) opposes the applied field (second term on the righthand side of (II-12)) to produce a small output field (II-12,(II-15). This phenomenon, due to collective response to an external field, is somewhat analogous to the Meisner effect (28) in the theory of superconductivity. In this regime the macroscopic transverse dipole moment, v, (II-17) is small, again due to the relatively small Stark shift.

Once the first turning point is reached (point A of curve a), by increasing y, there are no steady state values available for the system if y is further increased, unless the system jumps to a higher value of the output (point B of curve a). This jump

corresponds to a sudden decrease in g, the atom-atom coupling, and a sudden increase in the macroscopic polarization, v. Further increase in y produces a nearly linear, empty cavity response in x. That is, the material is nearly transparent to the input y, i.e., $x \approx y$. In this regime, the atom-atom pair correlation is destroyed and each individual atom's dipole moment follows the driving field y, coherently with definite phase. This could be called the regime of "cw self-induced transparency".



Fig. 1. Effect of cavity and laser detuning on optical bistability. Input field y vs. internal field x from (II-19). Curve a: c = 40, $\delta = v = 0$; curve b: c = 40, $\delta = v = 2$; curve c: c = 40, $\delta = v = 4$. $\delta = (\omega - \omega_0)/\gamma$; $v = (\varepsilon - \omega_0)/\gamma$; T = 0. Fig. 2. Asymmetric effect of cavity detuning on optical bistability. Input field y vs. internal field x from (II-19). Curve a: c = 60, $\delta = v = 10$; curve b: same parameters as for curve a, except $\delta = -9$; curve c: same parameters as for curve a, except $\delta = 9$; curve d: same parameters as for curve a, except $\delta = 8$; $T_a = 0$.

The asymmetric effects of cavity offtuning for fixed laser frequency is shown in Figure 2. These results exhibit large dispersive contributions for cavity offtuning and are in good qualitative agreement with the experimental results for sodium vapor (5).

The effective temperature dependence of the hysteresis cycle and OB threshold is shown in Figure 3 for input y vs. output x from (II-19) in units of γ . For convenience, we have taken perfect tuning, so (II-19) becomes, in units of γ ,

$$y = x + 2c \frac{x}{(1+x)^2} \tanh \frac{1}{2} \Gamma x$$
, (III-3)

where

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$$\Gamma = \gamma \beta_s = \gamma / k T_s$$
 (III-4)

B. Effective Temperature-induced Bistability and Hysteresis and the Optical Meisner Effect

It is interesting to consider the inversion of (III-3) to obtain Γ in terms of the input y and output x, (29),

$$\Gamma = \frac{1}{x} \ln \left\{ \frac{2cx + (y-x)(x^2+1)}{2cx - (y-x)(x^2+1)} \right\} .$$
 (III-5)

Equation (III-5) is plotted in Figure 4 for the inverse temperature, Γ vs. output field x for fixed y in units of γ . Each curve corresponds to a different value for the fixed input $y = y_c$. It is seen that bistability and hysteresis occur in the output field x by varying the inverse temperature Γ when the input y is fixed at a value above a certain threshold.

Consider the curve in Figure 4 showing the largest hysteresis area, corresponding to the appropriate fixed input field $y = y_c$. As the effective temperature T_s is increased (Γ decreased) from some small value to larger values, the output x increases slightly as the reaction field \bar{g}_V in (II-12), due to the collective atomic pair interaction which opposes the applied field α , is diminished as the temperature T_s is increased, (II-17). This is due to simultaneous reduction of the macroscopic dipole moment (II-17) because of the increase in effective temperature T_s , and to the decrease in \tilde{g} (II-8) as a consequence of the increase in the field-induced Stark shift, (II-10), (II-15).

As Γ is decreased to the value corresponding to point A in Figure 4, any further decrease in Γ will necessarily correspond to a stationary state on the upper part of the curve, point B. The system undergoes an abrupt increase in the output x as a consequence of the catastrophic breakdown of the atomic pair correlation and, hence, the reaction field \bar{g}_{ν} (II-12). In other words, along the path AB, Figure 4, the macroscopic transverse polarization ν (II-17) increases abruptly due to the increased Stark shift caused by the transition to

larger total local field. In the same process, however, the atomic pair correlation \overline{g} undergoes a catastrophic reduction in the transition from A to B. As Γ is further decreased, the macroscopic polarization diminishes due to the increase in T_S and the system approaches the state of atomic saturation (bleaching) at high absolute temperature. The reverse process takes place as Γ is increased (T_S decreased).





Fig. 3. Normalized externallyapplied field y vs. normalized internal field x according to Eq. (III-3) with c = 60. Each curve corresponds to a different value for Γ , and in order of decreasing values for the threshold value for y, $\Gamma = 1.00$, 0.65, 0.35, 0.25. The threshold value of Γ for bistability in this case is $\Gamma = 0.35$. The dotted lines and arrows indicate a particular hysteresis cycle.

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Fig. 4. Γ vs. x/y_c according to Eq. (III-5) for c = 60 and various values for the fixed externally-applied field y_c. In order of decreasing values for the threshold for Γ , y_c = 21.8, 21.7, 21.6, 21.3, 21.0, 20.0. The threshold value of y_c for bistability is y_c = 21.0 which is seen to be consistent with the threshold value of Γ for bistability in Fig. 3. The dotted lines and arrows indicate a particular hysteresis cycle.

The reduction of the internal field x due to atomic pair correlation (11-12), and the abrupt reduction of the reaction field at a critical temperature, T_s , with consequent increase in the total internal field x, for fixed input y_c shown in Figure 4 is quite analogous to the Meisner effect in the theory of superconductivity (15,28). We are tempted to label this effect, the "optical Meisner effect".

C. Second-order Phase Transition in the Absence of Applied Field

In the absence of applied field, i.e., for y = 0, (II-19) predicts the existence of a second-order phase transition, provided that the following gap equation is satisfied (12),

$$\tanh \frac{1}{2}\beta_{c}\varepsilon = \frac{\varepsilon}{2g_{c}^{2}} \frac{(\omega-\varepsilon)^{2}+\gamma^{2}}{\omega-\varepsilon}$$
, (III-6)

where T_c is here the actual temperature of the heat bath.

If this equation can be satisfied, it defines a critical temperature T_c below which the system exhibits a macroscopic polarization (ordered phase) and above which there is no macroscopic polarization (disordered phase). It is readily seen that (III-6) cannot be satisfied without offtuning provided by cavity mirrors. Furthermore, it is readily determined that for conditions where OB is observed in the optical regime, i.e., c > 4, (III-2), (III-6) will never be satisfied (12). However, it can be satisfied for microwave frequencies. This explains why this effect has not yet been observed in OB experiments.

IV. SUMMARY AND CONCLUSIONS

The effort at MICOM in quantum optics has produced results which for the first time predict OB which is caused by cooperative atom-atom interaction via their mutual radiation fields (12). The mechanism for the atomic pair correlation can be interpreted as due to virtual photon exchange interaction in analogy with the interaction between Cooper pairs via the phonon field in the BCS theory of superconductivity (30). The measure of the strength of the atomic pair correlation is \overline{g} (II-8). The main result of our theoretical model is the equation of state (II-19) relating the input field y to the output field x as a function of the inverse of the effective temperature $\beta_{\rm S}$.

The same mechanism is well known to be responsible for the cooperative atomic interaction which results in the dynamical process of superfluorescence (23,31). Our model also connects for the first

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time OB, superfluorescence of the superradiant phase transition in thermodynamic equilibrium in the absence of applied field (32).

A completely new and heretofore unpredicted phenomenon (29) is shown in Figure 4. Here, we have depicted the variation of the output field amplitude x as a function of the inverse temperature Γ for various input fields y_c from (III-5). These curves show bistability and reversible hysteresis in the output field x with variation of the inverse temperature Γ for fixed input field, y_c for the latter greater than a well-defined threshold value. These results could have important applications as a sensitive FIR, IR or millimeter wavelength detector with frequency upconversion. That is, an FIR or IR active material could be introduced as a buffer with the optically nonlinear material in a cavity. The FIR or IR energy absorbed by the buffer can be kinetically transferred to the optically nonlinear material via rotational-vibrational, translational energy transfer to alter Γ . Switching times in the hysteresis cycle could be on the order of the thermal fluctuation times for the system.

We also predict for the first time OB in a small volume without mirrors. For the case without mirrors, the mode frequency $\omega = \varepsilon$ and $\gamma = c/2L$ where L is the length of a cylindrical volume of the active material. The conditions for a cavity are that the mode frequency ω is the cavity frequency and need not equal the atomic frequency ε , and the photon escape rate $\gamma = c/2L\tau$ where τ is the transmission coefficient for the cavity mirrors. Optical bistability in a small volume without mirrors could have important applications for micron size OB elements imbedded in a neutral binder to constitute a two-dimensional optical digital memory bank or optical digital imaging surface. This certainly would alleviate the problem of having to produce highly uniform reflective surfaces on miniaturized OB elements. It may be also possible to constitute a three-dimensional optical digital memory bank of relatively compact dimensions.

Further work is in progress (27,33) to calculate the relevant switching rates in terms of material parameters and to determine the effects of atomic line shape on cooperative OB. We are also investigating the effects of multimode coupling to the Stark shifted atomic line shape. It is anticipated that this may lead to the establishment of regenerative oscillations which may have applications as passive high intensity laser cw to pulse conversion. We feel that our further work will undoubtedly lead to even further progress and new ideas.

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