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Intrinsic optical bistability in collections of spatially distributed two-level atoms

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We use a quantum-electrodynamical, many-body treatment to show mirrorless optical bistability in terms of the spatial properties of coherent dipole-dipole interactions among interacting two-level atoms. The general theory is applied to two special cases: (1) a thin sample of two-level atoms, with a width smaller than a resonance wavelength and (2) a long sample of two-level atoms, with dimensions very large relative to a resonance wavelength. While for the thin sample we are able to use a mean-field approximation with validity, for the long sample we are compelled to take into account retardation and propagation. In both cases bistability is found to be related to a renormalization of the frequency (or relaxation rate) that is inversion dependent. For the long sample the frequency renormalization is significant for high atomic densities and for large oscillator strengths.

I. INTRODUCTION

Intrinsic optical bistability (IOB) that is not caused by external feedback such as mirrors, has been the subject of recent intense interest.[1] It was first pointed out by Bowden and Sung[2(a)] and by Bowden[2(b)] that optical bistability (OB) may occur for a system comprised of a collection of atoms interacting with the electromagnetic field and driven by an externally applied coherent field without external feedback. The first detailed experimental study of intrinsic optical bistability was conducted almost simultaneously by Hajto and Janossy[3] using amorphous $GeSe_2$, who interpreted their results as due to temperature-dependent-induced optical absorption in the material, and Bohnert, Kalt, and Klingshirn,[4] who used CdS, and also Rossmann, Henneberger, and Voigt[5] using the same material. The process in the first case depends upon absorption due to temperature variation induced by the incident field, whereas the latter cases depend upon saturation of absorption due to the generation of carriers in the material, and the IOB has been interpreted[6] as due to band-gap renormalization. Since the earlier works, there have been many theoretical and experimental investigations of various forms of IOB.[1]

We present here a fully quantum-mechanical treatment of mirrorless (intrinsic) optical bistability (IOB) from a collection of a large number of spatially distributed two-level atoms interacting via the electromagnetic field and driven by an externally applied coherent field. Atomic cooperative effects in IOB have been treated in previous works either by assuming a small volume with dimensions smaller than a wavelength,[2,7] or by assuming a system with a small number of atoms in a semiclassical approximation.[8] In the present work we treat the problem from the many-body standpoint by developing the Heisenberg equations of motion in the "bad-cavity" limit, in which the variables associated with the field modes are adiabatically eliminated.[9]

The general theory is developed in Sec. II. We distinguish between terms in the equations of "spontaneous," "cooperative," "induced," and "Langevin force" origin. While the spontaneous, induced, and Langevin force terms depends only on the single atom response, the cooperative terms include factors of operators belonging to pairs of atoms with arguments expressive of retardation. In the cooperative terms the explicit spatial dependence of the dipole-dipole interactions enters in the coefficients $B(i,j)$ which turn out to be identical to those obtained from classical dipole-dipole interactions.[10,11]

We treat the steady-state conditions for our system and by taking the expectation values in the Heisenberg equations of motion, the Langevin force terms vanish. In the present work we do not treat fluctuations and the effects of quantum fluctuations in our system will be presented in a separate work.[12]

It has been established in various works[8,13–15] that factorization of the cooperative terms in steady-state results in a cubic nonlinearity and bistability. After establishing that factorization of different atomic operators is a suitable approximation for the distributed many-atom system we show bistability effects under steady-state conditions for two cases: (1) the thin sample geometry with propagation length smaller than a resonance wavelength and (2) the extended sample geometry where propagation effects are important.

In Sec. III we treat the thin sample in the mean-field approximation in which we use average values for the expectation values of the atomic operators. The present mean-field approximation ignores propagation effects and this is justified for a thin sample with a propagation distance smaller than a wavelength. For the steady-state conditions a cubic equation is derived which shows bistability in the parameter space. The linear stability analysis for this system is described in Sec. IV.

In Sec. V we treat a long sample by taking into account retardation and propagation. We analyze the cooperative effects and find that under appropriate approximations the conventional Maxwell-Bloch equations are reproduced but with a correction that can be expressed as an inversion-dependent renormalization of the frequency. This correction is proportional to the number of two-level atoms per cubic wavelength and to the decay constant, and therefore should be important for high densities and strong oscillator strengths. By taking into account propagation and retardation effects in the long sample we find the interesting result that the two phases, of high and low transmissivity, may coexist spatially in the material.

II. GENERAL THEORY

Our system is composed of a large number of spatially distributed two-level atoms coupled to each other only via the electromagnetic field and driven externally by an applied radiation field taken to be in a coherent state and propagating along the z axis with a linear polarization in the x direction. The Hamiltonian which describes the system is given in the rotating-wave and in the electric dipole approximations by[16,17]

$$H = H_0 + H' ,$$
$$H_0 = \tfrac{1}{2}\hbar\omega \sum_{i=1}^{N} \sigma_z^{(i)} + \hbar \sum_k \omega_k a_k^\dagger a_k , \tag{1}$$
$$H' = -i\hbar \sum_k \sum_{i=1}^{N} g_k^{(i)} a_k \sigma_+^{(i)} e^{i\mathbf{k}\cdot\mathbf{r}_i} - i\hbar/2 \sum_{i=1}^{N} \omega_R \sigma_+^{(i)} e^{-i(\omega_0 t - \mathbf{k}_0\cdot\mathbf{r}_i)} + \text{H.c.} ,$$

where H.c. denotes the Hermitian conjugate and

$$g_k^{(i)} = \left[\frac{2\pi\omega_k}{\hbar V} \right]^{1/2} P \hat{\mathbf{p}}_i \cdot \hat{\mathbf{x}} .$$

H_0 includes the free atoms and free-field Hamiltonian. We use here the usual operators related to the SU(2) algebra for a single atom: $\sigma_z^{(i)}$ represents the population-inversion operator and $\sigma_\pm^{(i)}$ are the raising and lowering operators for atom i with coordinates $\mathbf{r}_i$. ω_R is the Rabi rate associated with the applied coherent field. ω_0 and k_0 are the carrier frequency and wave vector of the applied field, respectively. V is the quantization volume for the field, ω_k is the frequency of the mode k, $\hat{\mathbf{x}}$ is a unit vector in the direction of polarization, P is the dipole moment matrix element, and $\hat{\mathbf{p}}_i$ is the unit vector for the dipole of atom i.

The Heisenberg equations of motion for our system are obtained in the bad-cavity limit by adiabatically eliminating the variables associated with the field modes.[9] We eliminate the rapid time and spatial dependence of the dipole operators by substituting

$$\sigma_{+0}^{(i)}(t) = \sigma_+^{(i)}(t) e^{-i(\omega_0 t - \mathbf{k}_0\cdot\mathbf{r}_i)} . \tag{2}$$

We get

$$\frac{d\sigma_z^{(i)}(t)}{dt} = -\beta_1[\sigma_z^{(i)}(t)+1] - \left[2\sum_{j=1}^{N}{}' B^*(i,j)\sigma_{+0}^{(i)}(t)\sigma_{-0}^{(j)}\left[t-\frac{r_{ij}}{c}\right] e^{i\mathbf{k}_0\cdot(\mathbf{r}_i-\mathbf{r}_j)} + \omega_R\sigma_{+0}^{(i)}(t) + 2\sigma_{+0}^{(i)}(t) f^{(+)}(\mathbf{r}_i,t) + \text{H.c.} \right] , \tag{3}$$

$$\frac{d\sigma_{+0}^{(i)}(t)}{dt} = (i\omega-\beta_2)\sigma_{+0}^{(i)}(t) + \sum_{j=1}^{N}{}' B(i,j)\sigma_{+0}^{(j)}\left[t-\frac{r_{ij}}{c}\right]\sigma_z^{(i)}(t) e^{-i\mathbf{k}_0\cdot(\mathbf{r}_i-\mathbf{r}_j)} + \frac{\omega_R^*}{2}\sigma_z^{(i)} + f^{(-)}(\mathbf{r}_i,t)\sigma_z^{(i)}(t) . \tag{4}$$

The prime on $\sum$ indicates $j \neq i$, and $f^{(+)}(\mathbf{r}_i,t)$ and $f^{(-)}(\mathbf{r}_i,t)$ are Langevin force operators which stem from the vacuum contribution to the fluctuations due to normal ordering of the field operators relative to the atomic operators in products of field and atomic operators in the equations of motion. These operators are δ correlated,

$$\langle f^{(+)}(r_i,t), f^{(-)}(r_i,t') \rangle \propto \delta(t-t') . \tag{5}$$

In our fully quantum-mechanical model $\beta_1 = 2\beta$, $\beta_2 = \beta$, where $\beta = 2|P|^2 k^3/3\hbar$ is defined as half the spontaneous decay constant. If we introduce additional homogeneous broadening, β_1 and β_2 may be considered as empirical constants.[12] In such an approach the model becomes less rigorous, but more general.

The coefficients $B(i,j)$, derived from the model, are identical to those obtained from classical dipole-dipole interactions:[11]

$$B(i,j) = \tfrac{3}{2}\beta\{[\hat{\mathbf{p}}_i\cdot\hat{\mathbf{p}}_j - (\hat{\mathbf{p}}_i\cdot\hat{\mathbf{r}}_{ij})(\hat{\mathbf{p}}_j\cdot\hat{\mathbf{r}}_{ij})]F_{\text{I}}(kr_{ij}) + (\hat{\mathbf{p}}_i\cdot\hat{\mathbf{r}}_{ij})(\hat{\mathbf{p}}_j\cdot\hat{\mathbf{r}}_{ij})F_{\text{II}}(kr_{ij})\} , \tag{6}$$

where

$$F_{\text{I}}(kr_{ij}) = e^{ikr_{ij}}(1/k^2r_{ij}^2 + i/k^3r_{ij}^3 - i/kr_{ij}) ,$$
$$F_{\text{II}}(kr_{ij}) = e^{ikr_{ij}}(-2i/k^3r_{ij}^3 - 2/k^2r_{ij}^2) . \tag{7}$$

Here $k = \omega/c$, $\hat{\mathbf{p}}_i$ and $\hat{\mathbf{p}}_j$ are the unit vectors for the dipoles of the atoms i and j, $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$ and $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$ is the distance between the two atoms.

In developing Eqs. (3) and (4) we sum the interaction of atom i with many other atoms described by the summation over j ($j \neq i$). In the present work we assume a model based upon many-body interactions in which each atom is influenced by the ensemble average over all other atoms. Such an assumption is justified when the number of atoms within a volume of a wavelength is large ($\gg 1$). We apply the equations either for steady-state conditions or for small fluctuations near the steady state (i.e., linear stability analysis). Under such conditions any correlation which is generated between the atoms due to initial conditions (like that assumed in the transient behavior of superradiance) is destroyed by the dephasing mechanism. The time of decorrelation is of order $1/\beta$ where β is the spontaneous decay time and becomes shorter if we add additional homogeneous broadening due to collisions. Under such conditions, as shown recently by Hopf and Bowden[8] in numerical simulations for a finite number of atoms within a volume of a wavelength, we can adequately justify factorization of the products of the dipole operators among different atoms. Factorization is further validated by the fact that there is a distribution of retardation times. Therefore we use the approximation

$$\langle \sigma_{+0}^{(i)}(t)\sigma_{-0}^{(j)}(t-r_{ij}/c)\rangle = \langle \sigma_{+0}^{(i)}(t)\rangle\langle \sigma_{-0}^{(j)}(t-r_{ij}/c)\rangle \;,$$
$$\langle \sigma_{+0}^{(j)}(t-r_{ij}/c)\sigma_{z}^{(i)}(t)\rangle = \langle \sigma_{+0}^{(j)}(t-r_{ij}/c)\rangle\langle \sigma_{z}^{(i)}(t)\rangle \;, \quad (8)$$

etc. By taking into account that expectation values of Langevin force terms vanish, [Eq. (4)], we get

$$\frac{d\langle \sigma_z(t)\rangle^{(i)}}{dt} = -\beta_1[\langle \sigma_z^{(i)}(t)\rangle + 1] - \left[\omega_R\langle \sigma_{+0}^{(i)}(t)\rangle + 2\sum_{j=1}^{N}{}' B^*(i,j)e^{i\mathbf{k}_0\cdot(\mathbf{r}_i-\mathbf{r}_j)}\langle \sigma_{+0}^{(i)}(t)\rangle\langle \sigma_{-0}^{(j)}(t-r_{ij}/c)\rangle + \text{c.c.}\right] \;, \quad (9)$$

$$\frac{d\langle \sigma_{+0}^{(i)}(t)\rangle}{dt} = (i\Delta-\beta_2)\langle \sigma_{+0}^{(i)}(t)\rangle + (\omega_R^*/2)\langle \sigma_z^{(i)}(t)\rangle + \sum_{j=1}^{N}{}' B(i,j)e^{-i\mathbf{k}_0\cdot(\mathbf{r}_i-\mathbf{r}_j)}\langle \sigma_{+0}^{(j)}(t-r_{ij}/c)\rangle\langle \sigma_z^{(i)}(t)\rangle \;, \quad (10)$$

where c.c. denotes the complex conjugate and $\Delta=\omega-\omega_0$ is the deviation of the applied field frequency from resonance. The reaction field is defined as

$$\Theta(r_i) \equiv \sum_{j=1}^{N}{}' B(i,j)e^{-i\mathbf{k}_0\cdot(\mathbf{r}_i-\mathbf{r}_j)}\langle \sigma_{+0}^{(j)}(t-r_{ij}/c)\rangle \;. \quad (11)$$

The cooperative terms in Eqs. (9) and (10) represent the effect of the reaction field, which for low values of the externally applied field, reduces the internal field. For higher values of the external field its value decreases suddenly and thus, as shown below, produces a first-order phase transition. In order to illuminate the physical properties of our system we apply our general equations to two special cases: (1) a thin sample of two-level atoms, with a width smaller than a resonance wavelength λ and (2) a long sample of two-level atoms, with dimensions very large relative to a wavelength.

III. THIN SAMPLE

For coherent radiation impinging on a thin film of two-level atoms with a large surface area and a width smaller than a wavelength λ we ignore the time of retardation r_{ij}/c by a Taylor expansion of the operators and use the approximation

$$e^{i\mathbf{k}_0\cdot(\mathbf{r}_i-\mathbf{r}_j)} \simeq 1 \;. \quad (12)$$

Under these approximations we can use mean values for the expectation values of the atomic operators:

$$\langle \sigma_{+0}^{(j)}(t-r_{ij}/c)\rangle \simeq \langle \sigma_{+0}^{(j)}(t)\rangle \simeq \langle \sigma_{+0}^{(i)}(t)\rangle = \langle \sigma_{+0}(t)\rangle \;,$$
$$\langle \sigma_{-0}^{(j)}(t-r_{ij}/c)\rangle \simeq \langle \sigma_{-0}^{(j)}(t)\rangle \simeq \langle \sigma_{-0}^{(i)}(t)\rangle = \langle \sigma_{-0}(t)\rangle \;. \quad (13)$$

After using Eqs. (12) and (13) in Eqs. (9) and (10), we get

$$\frac{d\langle \sigma_z\rangle}{dt} = -\beta_1(\langle \sigma_z\rangle + 1) - 4\,\mathrm{Re}\Gamma\langle \sigma_{+0}\rangle\langle \sigma_{-0}\rangle - \omega_R\langle \sigma_{+0}\rangle - \omega_R^*\langle \sigma_{-0}\rangle \;. \quad (14)$$

$$\frac{d\langle \sigma_{+0}\rangle}{dt} = (i\Delta-\beta_2)\langle \sigma_{+0}\rangle + \Gamma\langle \sigma_{+0}\rangle\langle \sigma_z\rangle + \frac{\omega_R^*}{2}\langle \sigma_z\rangle \;, \quad (15)$$

where all expectation values are taken at the same time t and

$$\Gamma = \sum_{j=1}^{N}{}' B(i,j) \;.$$

The mean-field approximation [Eq. (13)] becomes a very good approximation when Γ has only a small dependence on the location of each atom i in the thin sample. In such cases one can use Eqs. (14) and (15) with some mean value for Γ. In order to check the validity of this approximation and also in order to give some estimates for the value of Γ we have made the following calculation.

We assume that the thin sample is a cylindrical slab with a thickness $d \ll \lambda$. The z direction is defined as the direction $\hat{\mathbf{k}}$ of propagation of the external field and the dipoles induced by the externally applied field are assumed to be oriented along the $\hat{\mathbf{x}}$ axis. As described in Appendix A we calculate Γ as a function of z_i where $(0,0,z_i)$ is the location of the atom i and where the origin of the coordinates is assumed to be at the center of the cylinder. The general result represented in (A6) is quite complicated but for the limit $kd\to 0$, $k\epsilon\to 0$, $d/\epsilon=$const, we get a simple analytical result:

$$\Gamma = \frac{-3i\beta n\lambda^3}{8\pi^2}\left[1+\ln\frac{(d^2/4-z_i^2)^{1/2}}{\epsilon}\right] \;, \quad (16)$$

for

$$-d/2+\epsilon \le z_i \le d/2-\epsilon \;,$$

where n is the density of the two-level atoms. In the calculation we excluded a volume $\frac{4}{3}\pi\epsilon^3 = 1/n$ about $\mathbf{r}_i$ from the integration range in order to exclude the self-field of atom i. Γ depends on z_i only through the logarithmic function which is a slowly varying function. This conclusion is quite good even for atoms which are very near the surface [based on the comparison between Eqs. (A7) and (A8)].

For the thin film the main contribution to the reaction field comes from the dipole field in the "near region."[10] If the system has dimensions larger than a wavelength and if the dipole-dipole interaction is of spherical or cubic symmetry the average contribution of the dipole-dipole interaction to the near region vanishes. This is similar to the condition in the usual derivation of the Lorentz-Lorenz correction.[10] However, for a thin film with a width much smaller than a wavelength this local symmetry is broken due to the width of the film, and the contribution of the near region field becomes dominant. We find that in this case the value of Γ diverges unless we exclude the self-field from the volume of integration.

Under the steady-state conditions we get from Eqs. (14) and (15):

$$\langle\sigma_{+0}\rangle=\frac{-(\omega_R^*/2)\langle\sigma_z\rangle}{(i\Delta-\beta_2)+\Gamma\langle\sigma_z\rangle}\,, \tag{17}$$

$$\beta_1(\langle\sigma_z\rangle+1)[(\langle\sigma_z\rangle\mathrm{Re}\Gamma-\beta_2)^2+(\langle\sigma_z\rangle\mathrm{Im}\Gamma+\Delta)^2] = -\beta_2\,|\,\omega_R\,|^2\langle\sigma_z\rangle\;. \tag{18}$$

Equation (18) is a cubic equation in the population inversion and as proved in the next section, it leads to bistability effects. The critical points for $\langle\sigma_z\rangle$ as a function of $|\,\omega_R\,|^2$ are determined by taking the derivative of Eq. (18) with respect to $\langle\sigma_z\rangle$ and setting $d\,|\,\omega_R\,|^2/d\langle\sigma_z\rangle=0$. We get

$$\langle\sigma_z\rangle^2+F\langle\sigma_z\rangle+G=0\,, \tag{19}$$

$$F=\frac{2}{3}-\frac{\frac{4}{3}\mathrm{Re}(\Gamma)\beta_2}{|\,\Gamma\,|^2}+\left[\frac{4}{3}\right]\frac{\mathrm{Im}(\Gamma)\Delta}{|\,\Gamma\,|^2}\,, \tag{20}$$

$$G=\frac{[(\Delta^2+\beta_2^2)-2\,\mathrm{Re}(\Gamma)\beta_2+2\,\mathrm{Im}(\Gamma)\Delta+(\beta_2/\beta_1)\,|\,\omega_R\,|^2]}{3\,|\,\Gamma\,|^2}. \tag{21}$$

This must be satisfied simultaneously with Eq. (18). The condition for the threshold is $G=\frac{1}{4}F^2$ and by assuming the approximations $\Gamma\gg\beta_2$, $\Gamma\gg|\,\Delta\,|$, we get

$$\frac{\beta_2\,|\,\omega_R\,|^2}{\beta_1\,|\,\Gamma\,|^2}=\frac{1}{3} \tag{22}$$

for the threshold condition.

We find that, as the field intensity increases and the Rabi parameter becomes large relative to the cooperative constant Γ, the two-level system switches suddenly from the low-transmission branch to the high-transmission branch. We find here the interesting result that the switching occurs at relatively lower intensities if dephasing mechanisms due to collisions are introduced. This is illustrated in the next section in Fig. 4.

In previous works,[1,2,7,8] it was shown that nonlinear renormalization of the frequency may lead to bistability. In the present case such renormalization is obtained from Eqs. (14) and (15) in which $\Delta\to\Delta+\langle\sigma_z\rangle\mathrm{Im}(\Gamma)$.

According to the theoretical calculations made in Appendix A, Γ becomes imaginary in the limit that $kd\to 0$, $d/\epsilon=$const. By performing numerical calculations of Γ for a thin sample, we found that the real part of Γ was small relative to the imaginary part and that the ratio decreases as a function of the density of atoms. While the imaginary part of Γ leads to a renormalization of the frequency, the real part of Γ, which is positive, is related to superradiance. According to the present analysis both the real part and imaginary part of Γ may lead to bistability. In treating the bistability effect for the thin sample one should note that $\langle\sigma_{+0}\rangle$ is proportional to ω_R^* and it will tend to zero only for a vanishing external field amplitude.

IV. LINEAR STABILITY ANALYSIS FOR THE THIN SAMPLE

Here we show that the solutions to the steady-state conditions of Eq. (18) indeed exhibit bistability. Let us consider a stationary state of the thin sample. For infinitesimal perturbations of the system from the stationary state the linearized set of equations obtained from Eqs. (14) and (15) are

$$\begin{aligned}\frac{d\delta\langle\sigma_z\rangle}{dt}=&-\beta_1\delta\langle\sigma_z\rangle-4\,\mathrm{Re}\Gamma(\delta\langle\sigma_{+0}\rangle)\langle\sigma_{-0}\rangle\\&-4\,\mathrm{Re}\Gamma\langle\sigma_{+0}\rangle(\delta\langle\sigma_{-0}\rangle)\\&-\omega_R(\delta\langle\sigma_{+0}\rangle)-\omega_R^*\delta\langle\sigma_{-0}\rangle\,,\end{aligned} \tag{23}$$

$$\begin{aligned}\frac{d\delta\langle\sigma_{+0}\rangle}{dt}=&(i\Delta-\beta_2)\delta\langle\sigma_{+0}\rangle+\Gamma(\delta\langle\sigma_{+0}\rangle)\langle\sigma_z\rangle\\&+\Gamma\langle\sigma_{+0}\rangle(\delta\langle\sigma_z\rangle)+(\omega_R^*/2)\delta\langle\sigma_z\rangle\,,\end{aligned} \tag{24}$$

where $\langle\sigma_{+0}\rangle$ and $\langle\sigma_z\rangle$ are again given by Eqs. (17) and (18). The equation for $\delta\langle\sigma_{-0}\rangle$ is the complex conjugate of Eq. (24).

By considering the three-component vector $\mathbf{q}=(\delta\langle\sigma_z\rangle,\delta\langle\sigma_{+0}\rangle,\delta\langle\sigma_{-0}\rangle)$ and following a general procedure for linear stability analysis,[18] we introduce in the linearized equations the ansatz

$$\mathbf{q}(t)=\mathbf{q}(0)\exp(\lambda t)\,, \tag{25}$$

where λ is a complex number. We get the polynomial equation for the stability eigenvalues,

$$\lambda^3+A_1\lambda^2+A_2\lambda+A_3=0\,, \tag{26}$$

where

$$\begin{aligned}A_1=&\beta_1+2\beta_2-2\,\mathrm{Re}(\Gamma)\langle\sigma_z\rangle\,,\\A_2=&(\beta_1/\beta_2)(\langle\sigma_z\rangle+1)\mathrm{Im}(\Gamma)[\Delta+\mathrm{Im}(\Gamma)\langle\sigma_z\rangle]+(3\beta_1/\beta_2)\mathrm{Re}(\Gamma)(\langle\sigma_z\rangle+1)[\mathrm{Re}(\Gamma)\langle\sigma_z\rangle-\beta_2]\\&+[(\langle\sigma_z\rangle\mathrm{Re}\Gamma-\beta_2)^2+(\langle\sigma_z\rangle\mathrm{Im}\Gamma+\Delta)^2][1-(\beta_1/\beta_2)(\langle\sigma_z\rangle+1)/\langle\sigma_z\rangle]\\&+2\beta_1\beta_2-2\beta_1\mathrm{Re}(\Gamma)\langle\sigma_z\rangle-2(\mathrm{Re}\Gamma)^2(\beta_1/\beta_2)\langle\sigma_z\rangle(\langle\sigma_z\rangle+1)\,,\\A_3=&2\beta_1(\langle\sigma_z\rangle+1)[\,|\,\Gamma\,|^2\langle\sigma_z\rangle-\beta_2\mathrm{Re}(\Gamma)+\Delta\,\mathrm{Im}(\Gamma)]-(\beta_1/\langle\sigma_z\rangle)\{(\langle\sigma_z\rangle\mathrm{Re}\Gamma-\beta_2)^2+[\langle\sigma_z\rangle\mathrm{Im}(\Gamma)+\Delta]^2\}\;.\end{aligned} \tag{27}$$

In deriving Eqs. (27) we eliminated $\langle\sigma_{+0}\rangle$, $\langle\sigma_{-0}\rangle$, and $|\omega_R|^2$ by the use of Eqs. (17) and (18).

According to the Hurwitz criterion[19] the polynomial of Eq. (26) has only zeros with negative real parts if the three conditions

$$A_1>0,\ \ A_1A_2-A_3>0,\ \ A_3>0 \tag{28}$$

are fulfilled. These are therefore the conditions for our system to be stable under small shifts from a stationary state.

Since, according to Eq. (18), the equation for $\langle\sigma_z\rangle$ at the turning points is

$$\frac{-A_3}{\beta_2\langle\sigma_z\rangle}=0\ , \tag{29}$$

the condition of stability $A_3>0$ implies that the system is, as expected,[18] unstable in the region in which we have a negative slope:

$$d\langle\sigma_z\rangle/d\,|\omega_R|^2<0\ .$$

For positive values of Re(Γ) the condition $A_1>0$ is always fulfilled (as $\langle\sigma_z\rangle$ is, in our system, always negative). Our system becomes unstable in regions of positive slope[20] when Re(Γ) is negative and $2\,\mathrm{Re}(\Gamma)\langle\sigma_z\rangle>\beta_1+\beta_2$. Although in our theoretical and numerical calculations, Re(Γ) was positive, there might be cases of subradiance for which Re(Γ) is negative and the instabilities obtained in such cases should be of interest.

In order to illustrate the properties of the steady states in the thin sample we have calculated $\langle\sigma_z\rangle$ as a function of $|\omega_R|^2$ [Eq. (18)] and the stability conditions [Eqs. (28)] for various examples in which we vary the physical parameters. Some representative examples are pictured in Figs. 1–5 where the solid and dotted curves represent, respectively, stable and unstable steady states. We have taken $\beta_1=2$, so that all rates are essentially in units of $(\beta_1/2)$; thus $|\omega_R|^2$ is in units of $(\beta_1/2)^2$.

While in Figs. 1–4, Re(Γ)≥ 0 and the instabilities are only in the regions of a negative slope, in Fig. 5, Re(Γ)<0 and the instabilities are also in regions of a positive slope.[20] These figures demonstrate therefore the conditions $A_1>0$ and $A_3>0$ for the stability of our system. The condition $A_1A_2>A_3$ was fulfilled in all the numerical calculations in which $A_1>0$ and $A_3>0$ and therefore did not give additional instabilities.

According to the theoretical analysis the figures are invariant to simultaneous changes in the signs of Im(Γ) and Δ. As demonstrated in these figures the bistability is related to the imaginary and/or the real part of Γ.

In Figs. 1–3 and 5 we assumed the relation $\beta_2=\beta_1/2$ which follows from the fully quantum-mechanical model in which the spontaneous decay stems from the self-field

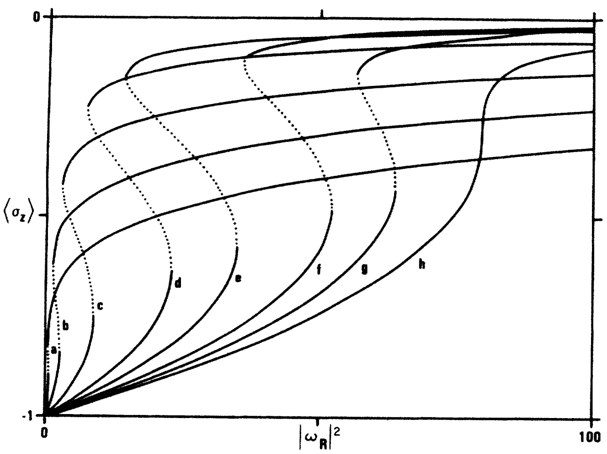

FIG. 2. $\langle\sigma_z\rangle$ as a function of $|\omega_R|^2$ for a thin sample with normalized parameters: $\beta_1=2$, $\beta_2=1$, Re(Γ)=0, Im(Γ)=−10. The deviation from resonance is given by (a) $\Delta=-8$, (b) $\Delta=-6$, (c) $\Delta=-4$, (d) $\Delta=-2$, (e) $\Delta=-1$, (f) $\Delta=0$, (g) $\Delta=\frac{1}{2}$, (h) $\Delta=1$. The solid and dotted curves represent, respectively, stable and unstable states.

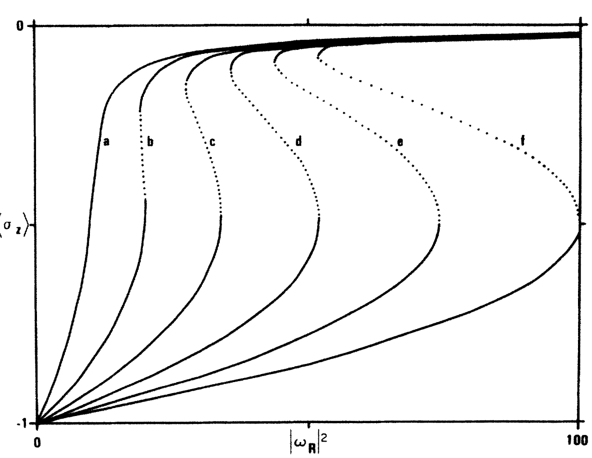

FIG. 1. $\langle\sigma_z\rangle$ as a function of $|\omega_R|^2$ for a thin sample with normalized parameters: $\beta_1=2$, $\beta_2=1$, $\Delta=0$, Re(Γ)=0. The imaginary value of Γ is given by (a) $\Gamma_I=-4$, (b) $\Gamma_I=-6$, (c) $\Gamma_I=-8$, (d) $\Gamma_I=-10$, (e) $\Gamma_I=-12$, (f) $\Gamma_I=-14$. The solid and dotted curves represent, respectively, stable and unstable states.

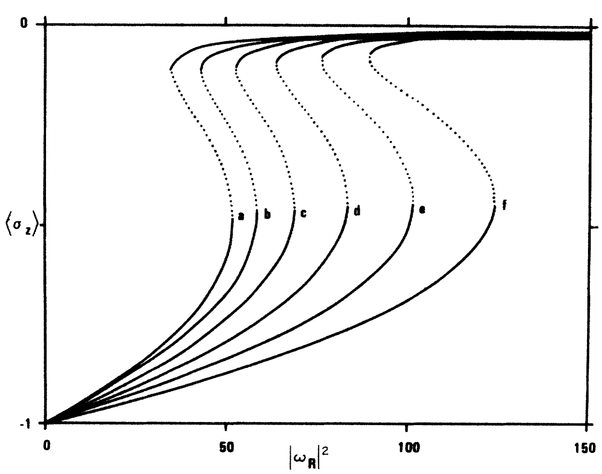

FIG. 3. $\langle\sigma_z\rangle$ as a function of $|\omega_R|^2$ for a thin sample with normalized parameters: $\beta_1=2$, $\beta_2=1$, $\Delta=0$, Im(Γ)=−10. The real part of Γ is given by (a) $\Gamma_R=0$, (b) $\Gamma_R=2$, (c) $\Gamma_R=4$, (d) $\Gamma_R=6$, (e) $\Gamma_R=8$, (f) $\Gamma_R=10$. The solid and dotted curves represent, respectively, stable and unstable states.

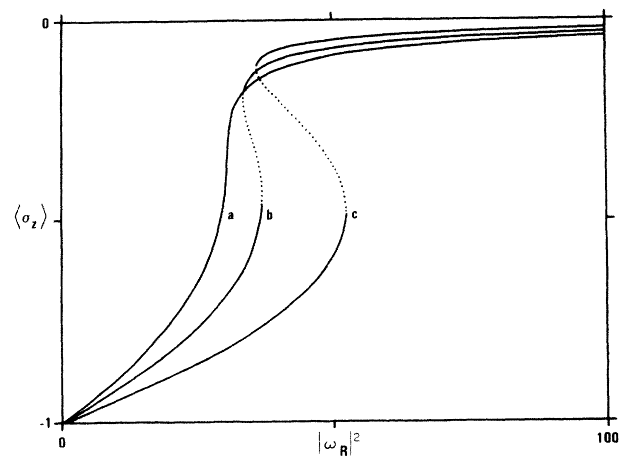

FIG. 4. $\langle \sigma_z \rangle$ as a function of $|\omega_R|^2$ for a thin sample with normalized parameters: $\beta_1=2$, $\Delta=0$, $\mathrm{Re}(\Gamma)=0$, $\mathrm{Im}(\Gamma)=-10$. The dephasing constant is given by (a) $\beta_2=2$, (b) $\beta_2=1.5$, (c) $\beta_2=1$.

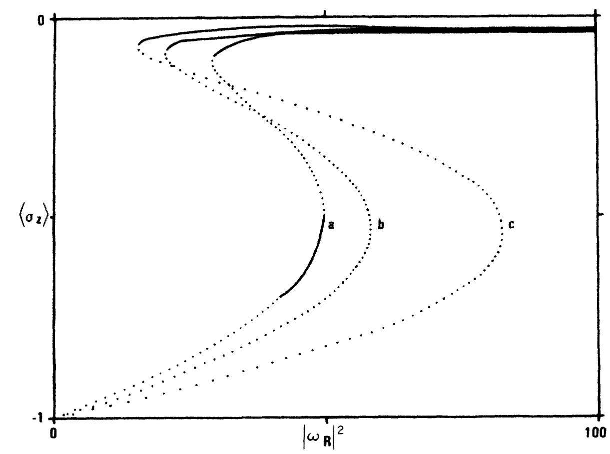

FIG. 5. $\langle \sigma_z \rangle$ as a function of $|\omega_R|^2$ for a thin sample with normalized parameters: $\beta_1=2$, $\beta_2=1$, $\Delta=0$, $\mathrm{Im}(\Gamma)=-10$. The real part of Γ is given by (a) $\Gamma_R=-2$, (b) $\Gamma_R=-6$, (c) $\Gamma_R=-10$. The solid and dotted curves represent, respectively, stable and unstable states.

part of the atom-field interaction. For taking into account additional homogeneous broadening, one may consider the ratio β_2/β_1 as an empirical parameter and the effect of changing this parameter is illustrated in Fig. 4.

The threshold condition given in Eq. (22) is in fairly good agreement with our numerical calculations. The explicit threshold condition for bistability in terms of $\mathrm{Im}(\Gamma)$ shown in Fig. 1 is consistent with similar threshold conditions determined in Refs. 7 and 8. Further conclusions on the qualitative behavior of our system can be obtained by examining the systematic dependence of the figures on the various system parameters.

V. LONG SAMPLE WITH RETARDATION AND PROPAGATION EFFECTS

For coherent radiation transmitted through a long sample, with dimensions very large relative to a wavelength, we use again Eqs. (9) and (10) and study the effect of the spatially varying parameters on the behavior of the system. We define the z axis as the direction of propagation of the externally applied field, and the x axis as the direction of polarization. Since the dipoles induced by the externally applied field are approximately in the x direction we simplify the expression given in Eq. (6) for $B(i,j)$ by assuming

$$\mathbf{p}_i=\mathbf{p}_j=P\hat{\mathbf{x}} \; . \tag{30}$$

For calculating the cooperative interaction of atom i with all other atoms we separate this interaction into two parts. We choose a sphere around the atom i with a radius r_0 that is on the order of a wavelength, so that within this sphere we can ignore retardation and use the mean-field approximation. The second part of the cooperative interaction includes the interaction with all other atoms that are not located inside the sphere. For the second part of the interaction we take into account retardation and propagation effects but simplify the calculations by using the approximation $r_{ij}>\lambda$, where r_{ij} is the distance between atom i and any other atom j that is outside the sphere. We transform the summations to integrations by using spatially continuous variables for the atomic expectation values. We calculate the total cooperative effect by adding the two parts of the interaction and the result, as expected, is found to be independent of our arbitrary choice of the radius r_0 of the sphere.

By using the mean-field approximation of Eq. (13) we express the first part of the cooperative interaction as

$$\begin{aligned} [d\langle \sigma_z^{(i)}(t)\rangle/dt]_{\mathrm{coop},(i)} &= -4\langle \sigma_{+0}^{(i)}(t)\rangle\langle \sigma_{-0}^{(i)}(t)\rangle \mathrm{Re}\Gamma_1 \; ,\\ [d\langle \sigma_{+0}^{(i)}(t)\rangle/dt]_{\mathrm{coop},(i)} &= \langle \sigma_{+0}^{(i)}(t)\rangle\langle \sigma_z^{(i)}(t)\rangle \Gamma_1 \; , \end{aligned} \tag{31}$$

where for $\mathbf{k}_0 \simeq \mathbf{k}=k\hat{\mathbf{z}}$ we get

$$\Gamma_1=\sum_j{}^{(i)} B(i,j) e^{-ik(z_i-z_j)} \; . \tag{32}$$

In Appendix B we calculate Γ_1 for the atom i which is at the center of a sphere with a radius r_0 and exchange the summation by integration for randomly distributed two-level atoms with a density n. In principle, one should eliminate the self-field by excluding from the integral a small sphere around atom i with a radius ϵ where $(4\pi/3)\epsilon^3 n=1$. Since the integrand is well behaved for small r (see Appendix B), we assume the approximation $\epsilon=0$ for $k\epsilon \ll 1$.

The final result for Γ_1 given by Eq. (B4) can be written as

$$\Gamma_1=\frac{3\beta n\lambda^3}{8\pi^2}\gamma \; , \tag{33}$$

$$\gamma=R-\tfrac{1}{2}\sin(2R)-\frac{1+\cos(2R)}{R}+\frac{2\sin(2R)}{R^2}-\frac{1-\cos(2R)}{R^3}-i\left[-\tfrac{7}{6}+\tfrac{1}{2}\cos(2R)-\frac{\sin(2R)}{R}-\frac{2\cos(2R)}{R^2}+\frac{\sin(2R)}{R^3}\right] , \tag{34}$$

where $R = kr_0$.

The real and imaginary parts of γ are described in Fig. 6 as functions of r_0/λ. Ignoring small oscillations we find that $\mathrm{Re}(\Gamma)$ is given approximately by a linearly increasing function of r_0,

$$\mathrm{Re}(\gamma) \simeq \frac{2\pi}{\lambda} r_0 = R \ . \tag{35}$$

As we increase the radius r_0 of the sphere, the contribution from the far dipoles becomes more dominant and the effect of the oscillatory part of $\mathrm{Re}(\gamma)$ becomes negligible.

$\mathrm{Im}(\gamma)$ is a negative oscillating function of r_0/λ with an averaged value

$$\mathrm{Im}(\gamma) = -\tfrac{7}{6} \ , \tag{36}$$

which is independent of r_0 for $r_0/\lambda > 1$. The imaginary part of γ is therefore contributed by the dipoles in the "intermediate region" and its effect on the equations of motion is to produce, through Eq. (31), a renormalization of the resonance frequency. According to Eq. (33), the frequency renormalization is proportional to the density of the two-level atoms and to the oscillator strength.

For calculating the second part of interaction with the distant dipoles which are outside of the sphere, we should take into account retardation and propagation effects. For this purpose it is convenient, at this stage of the calculation, to describe the expectation values of the atomic operators as continuous variables. We define $\langle \sigma_{+0}(\mathbf{r},t) \rangle$ and $\langle \sigma_z(\mathbf{r},t) \rangle$, respectively, as the complex dipole and the inversion of population per unit volume at point $\mathbf{r}$, consistent with the volume of integration of the sphere of radius r_0. Equations (9) and (10) can be written in the new variables as

$$\frac{d\langle \sigma_z(\mathbf{r},t)\rangle}{dt} = -\beta_1[\langle \sigma_z(\mathbf{r},t)\rangle + n] - \{[2\Theta^*(\mathbf{r},t) + \omega_R]\langle \sigma_{+0}(\mathbf{r},t)\rangle + \text{c.c.}\} \ , \tag{37}$$

$$\frac{d\langle \sigma_{+0}(\mathbf{r},t)\rangle}{dt} = (i\Delta - \beta_2)\langle \sigma_{+0}(\mathbf{r},t)\rangle + \left[\Theta(\mathbf{r},t) + \frac{\omega_R^*}{2}\right]\langle \sigma_z(\mathbf{r},t)\rangle \ , \tag{38}$$

where $\Theta(\mathbf{r},t)$ is a continuous variable corresponding to the definition of reaction field $\Theta(\mathbf{r}_i,t)$ given in Eq. (11), and is separated into two parts: $\Theta = \Theta_1 + \Theta_2$ corresponding to the contributions within and without the sphere. According to Eq. (31), we get

$$\Theta_1(\mathbf{r},t) = \langle \sigma_{+0}(\mathbf{r},t)\rangle \Gamma_1/n \ . \tag{39}$$

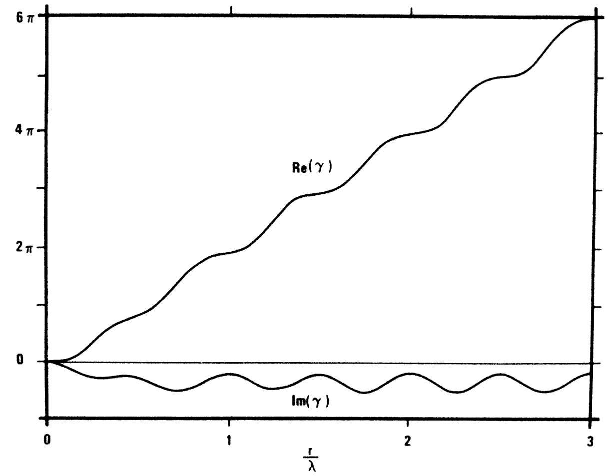

FIG. 6. $\mathrm{Re}(\gamma)$ and $\mathrm{Im}(\gamma)$ described as functions of r/λ.

For calculating the second part of the reaction field, we use the "radiation-zone approximation"[10] for $B(i,j)$, which is valid for $r_{ij} > \lambda$:

$$B(i,j) = -\frac{3i\beta}{2}\frac{e^{ikr_{ij}}}{kr_{ij}}\sin^2\alpha \ , \tag{40}$$

where α is the angle between $\mathbf{r}_{ij}$ and the x axis. The second part of Θ can be written as

$$\Theta_2(\mathbf{r},t) = -\frac{3i\beta}{2}\int' d^3r' \left\langle \sigma_{+0}\left[\mathbf{r}', t - \frac{|\mathbf{r}-\mathbf{r}'|}{c}\right]\right\rangle \times \frac{e^{-i\mathbf{k}_0\cdot(\mathbf{r}-\mathbf{r}')}e^{ik|\mathbf{r}-\mathbf{r}'|}}{k|\mathbf{r}-\mathbf{r}'|}\sin^2\alpha \ . \tag{41}$$

The prime on the integral indicates that we have to exclude from the volume of integration the sphere which includes the first part of the interaction.

For calculating $\Theta_2(\mathbf{r})$, we assume that the externally applied radiation is nearly resonant with the two-level atomic frequency so that $k \simeq k_0$, and find that, because of the spatial phase factors the main contribution to the integral comes from the region for which $\hat{\mathbf{k}}_0\cdot(\mathbf{r}-\mathbf{r}') \simeq |\mathbf{r}-\mathbf{r}'|$. The strength of the coherent dipole-dipole interaction for each atom is therefore mainly due to atoms that are located at smaller values of the propagation distance in the material. Following these considerations, we assume that the expectation values of the atomic operators per unit volume depend on the z coordinate and use in Eq. (41) the approximation $\sin^2\alpha = 1$, $|\mathbf{r}-\mathbf{r}'| \simeq |z-z'|$ (for the denominator) and $|y-y'| \ll |z-z'|$, $|x-x'| \ll |z-z'|$ (for the exponents). Following these approximations we get, by a straightforward integration of Eq. (41) over the x and y coordinates, the result

$$\Theta_2(r,t) = \Theta_2(z,t) = \frac{-3i\beta}{2}\int_0^{z-r_0} dz' \frac{\langle \sigma_{+0}(z', t-(z-z')/c)\rangle}{k(z-z')}\int\int dx\,dy\,\exp\{ik[(y-y')^2 + (x-x')^2]/2(z-z')\}$$
$$= \frac{3\pi\beta}{k^2}\int_0^{z-r_0} dz' \langle \sigma_{+0}(z', t-(z-z')/c)\rangle \ . \tag{42}$$

We find according to Eq. (42) that for each atom the dipole-dipole interaction with other atoms decreases as the inverse of distance but that due to phase factors appearing in the equations, the number of atoms in the opposite direction to that of propagation of the external field, which contribute to the cooperative effect, increases proportional to distance. So the explicit dependence of the cooperative dipole-dipole interaction on distance is completely eliminated.

The choice of r_0 defines the spherical volume over which Θ_1 is calculated. This volume must be larger than a cubic wavelength in order that the radiation-zone approximation be valid in the calculation for Θ_2. On the other hand, it should be small enough so that retardation effects are not important within this sphere. By using Eq. (36) we find that the average value of $\mathrm{Im}(\gamma)$ is independent of r for $r/\lambda > 1$. The effect of $\mathrm{Im}(\gamma)$, which enters in Eq. (39) via Eq. (33), is very important as it leads to a renormalization of the frequency. Following these considerations we define r_0 to be of order λ, but somewhat larger than λ

Concerning $\mathrm{Re}(\gamma)$ one should note that the effect of $\mathrm{Re}(\gamma)$ in Θ_1 is overwhelmed by the large value of Θ_2 in the long sample, since both terms influence the relaxation. Also by ignoring retardation in the small volume of radius r_0 and by neglecting the small oscillations of $\mathrm{Re}(\gamma)$ in Eq. (34) we find, according to Eqs. (31) and (39), that the net effect of $\mathrm{Re}(\gamma)$ is to allow the upper limit of integration for Θ_2 to become z instead of $z-r_0$.

We arrive now, after the above calculations, to an important physical result. Since $\langle \sigma_{+0}(z',t-(z-z')/c)\rangle$ of Eq. (42) is taken at retarded time the derivative $d\Theta_2/dz$ gives the form of Maxwell's equation in the slowly varying envelope approximation and in the retarded time frame. However, because of the macroscopic self-field contribution $\Theta_1(\mathbf{r})$ we should take into account in Eq. (38) frequency and dephasing renormalization that is proportional to the population inversion $\langle \sigma_z(\mathbf{r})\rangle$. This result is remarkable since in all textbooks on quantum optics in which Maxwell-Bloch equations are used, this effect is neglected. The present analysis shows that it may be important for high densities and large oscillator strengths.

In summary, the equation for $\Theta_2(z,t)$ is given by the form of the ordinary Maxwell equation in the slowly varying envelope approximation:

$$\frac{1}{c}\frac{\partial \Theta_2(z,t)}{\partial t}+\frac{\partial \Theta_2(z,t)}{\partial z}=\frac{3\pi\beta}{k^2}\langle \sigma_{+0}(z,t)\rangle \ , \qquad (43)$$

while in the Bloch equations represented by Eqs. (37) and (38) one should substitute $\Theta=\Theta_1+\Theta_2$, where Θ_1 leads to a renormalization of the frequency, relaxation, and dephasing.

In the present theory the spherical volume over which the discrete variables are averaged to obtain the continuous variables can be of order $r_0 > \lambda$. The use of the factorization and mean-field approximations for calculating Θ_1 is justified only for densities which are sufficiently large so that we have many atoms within a volume of a wavelength.

In previous works,[1,2,7,8] it has been shown that a renormalization of frequency can lead to bistability. In the present work we have clarified the nature of this effect in a many-atom system and found that the renormalization of frequency is proportional to the decay constant β multiplied by the number of two-level atoms per cubic volume of wavelength, consistent with the results of Refs. 7 and 8.

If one introduces in Eqs. (37) and (38) the definitions

$$\begin{aligned}\omega_R'(\mathbf{r})&=\omega_R+2\Theta_2^*(\mathbf{r})\ ,\\ \Gamma'&=\Theta_1(\mathbf{r})/\langle \sigma_{+0}(\mathbf{r})\rangle\\ &\simeq -i(3/8\pi^2)\tfrac{7}{6}\lambda^3\beta\ ,\end{aligned} \qquad (44)$$

these equations are of the same form as Eqs. (14) and (15) for the thin sample, with an imaginary value for Γ. The threshold condition for bistability represented in Eq. (22) will be correct here also when we exchange ω_R and Γ by ω_R' and Γ', respectively. In the long sample, $|\omega_R'(\mathbf{r})|$ will be a decreasing function of the distance along the z axis. We get therefore the important result that the two phases of high and low transmissivity may coexist spatially in the material.

VI. CONCLUSIONS

In the present work we have presented a general quantum-mechanical treatment of coherent dipole-dipole interactions in a coherently driven collection of a large number of spatially extended, two-level atoms, and have shown that a frequency renormalization (as well as relaxation renormalization) that is proportional to the population inversion exists and may lead to intrinsic optical bistability. In the context of the general theory we clarified by our analysis the relation between these effects and the use of the conventional Maxwell-Bloch equations.

We conclude from the present analysis that intrinsic (mirrorless) optical bistability may be produced by a thin sample of two-level atoms with a width smaller than a wavelength, due to a local spatial symmetry breaking. A system of a thin sample with two-level atoms might be realized in experiments with Rydberg atoms. While the present analysis is suitable mainly for an atomic system, it suggests that similar effects may be obtained by using crystals with a high density of bound excitons with large oscillator strengths. The treatment of the long sample shows the possibility that two phases, of high and low transmissivity, may coexist spatially in the material.

According to the analysis in the present article, the effects of intrinsic bistability follow mainly from the inversion-dependent frequency renormalization. For the long sample, this conclusion follows from Eqs. (37) and (38), with the value of $\Theta_1=\Gamma'\langle \sigma_{+0}\rangle$ given by Eq. (44) (where Γ' is imaginary). For the thin sample, it follows from Eqs. (17) and (18), with the value of Γ estimated in Eq. (A7) (the effect of relaxation renormalization expressed by the real part of Γ is negligible for high densities). In order to get the phase transition in the two cases, the frequency of the coherent radiation should be somewhat lower than the atomic frequency and of such a value that the frequency renormalization will bring it in and out of resonance as a function of the value of $\langle \sigma_z\rangle$. To obtain a good condition for intrinsic bistability, $\mathrm{Im}\Gamma$ should be made an order of magnitude (or more) larger than β_2, where β_2 is the relaxation rate for the complex dipole. Assuming for the pure quantum case $\beta_2=\beta$, where β is one-half of the spontaneous decay rate, we find that we need approximately 50–100 (or more) atoms within the volume of a cubic wavelength in order that the effects will be significant. If we introduce additional homogeneous broadening, which increases the value of β_2, the density should be increased by an additional factor of β_2/β. The present effects should therefore be significant for high densities and/or large oscillator strengths and when the mechanism of spontaneous decay is strong. The intensity of the external field for which the phase transition occurs can be estimated by (22).

In the present work we have used the mean-field approximation in the small volume to evaluate Θ_1, Eq. (39), which enables us to treat intrinsic bistability by an analytical quantum-mechanical calculation. The validity of the present model is supported by previous treatments of this problem from other points of view. The use of a semiclassical model[7] consisting of the Maxwell-Bloch formulation for a medium consisting of laser-driven two-level atoms with the local-field correction, where the mean-field and slowly varying envelope approximations were not made, gives precisely the same results for the renormalization of frequency and the same threshold conditions for bistability in the thin sample as the present work. The results of the heuristic model[8] which treats the intrinsic bistability and the dynamical statistical behavior by numerical simulations, for a finite number of atoms within a cubic wavelength quantitatively confirms the essential deterministic results derived in the present work, as well as that of Ref. 7. However, the present treatment is more general than the previous work, and is also analytical and therefore should be of much interest. We hope that experiments will be done for observing the phenomena predicted theoretically by the present analysis.

ACKNOWLEDGMENTS

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APPENDIX A: CALCULATION OF Γ FOR THE THIN SAMPLE

The present theoretical calculation is done by using the approximation (13) and assuming $\mathbf{k}_0\simeq\mathbf{k}=k\hat{\mathbf{z}}$. Locate atom i at $(0,0,z_i)$ and assume cylindrical symmetry, where the origin of coordinates is in the center of the cylinder. Then

$$\Gamma=\tfrac{3}{2}\beta n\int_0^{2\pi}d\phi\int_{-d/2}^{d/2}dz\int_{\rho_{\min}}^{\rho_{\max}}\rho\,d\rho[\sin^2\phi\,F_{\rm I}(kr)+\cos^2\phi\,F_{\rm II}(kr)]\ ,\tag{A1}$$

where $r=[\rho^2+(z-z_i)^2]^{1/2}$. In addition, we exclude a volume $(4/3)\pi\epsilon^3=1/n$ about $\mathbf{r}_i$ from the integration range. Then, performing the angle integration, we find

$$\Gamma=\tfrac{3}{2}\beta n\left[\int_{z_i-\epsilon}^{d/2}dz\int_0^{\rho_{\max}}\rho\,d\rho+\int_{-d/2}^{z_i-\epsilon}dz\int_0^{\rho_{\max}}\rho\,d\rho+\int_{z_i-\epsilon}^{z_i+\epsilon}dz\int_{[\epsilon^2-(z-z_i)^2]^{1/2}}^{\rho_{\max}}\rho\,d\rho\right][F_{\rm I}(kr)+F_{\rm II}(kr)]\ .\tag{A2}$$

Now,

$$\begin{aligned}\int\rho\,d\rho[F_{\rm I}(kr)+F_{\rm II}(kr)]&=\int r\,dr\,e^{ikr}(-i/kr-1/k^2r^2-i/k^3r^3)\\&=-1/k^2\int dR\,e^{iR}(i+1/R+i/R^2)\end{aligned}\tag{A3}$$

where $R\equiv kr$, so

$$\Gamma=-\frac{3}{2}\frac{\pi\beta n}{k^2}\left[\int_{z_i+\epsilon}^{d/2}dz\int_{k|z-z_i|}^{R_{\max}}dR+\int_{-d/2}^{z_i-\epsilon}dz\int_{k|z-z_i|}^{R_{\max}}dR+\int_{z_i-\epsilon}^{z_i+\epsilon}dz\int_{k\epsilon}^{R_{\max}}dR\right]e^{iR}(i+1/R+i/R^2)\tag{A4}$$

since

$$e^{iR}(i+1/R+i/R^2)=\partial/\partial R\,(e^{iR}-ie^{iR}/R)\ ,\tag{A5}$$

$$\begin{aligned}\Gamma=-\frac{3}{2}\frac{\pi\beta n}{k^3}&\left[k\int_{-d/2}^{d/2}dz\left[e^{iR_{\max}(z)}-\frac{ie^{iR_{\max}(z)}}{R_{\max}(z)}\right]\right.\\&\left.-2\epsilon k\left[e^{ik\epsilon}-\frac{ie^{ik\epsilon}}{k\epsilon}\right]-\left[\int_{z_i+\epsilon}^{d/2}dz+\int_{-d/2}^{z_i-\epsilon}dz\right]\left[ke^{ik|z-z_i|}-\frac{e^{ik|z-z_i|}}{|z-z_i|}\right]\right]\ ,\\R_{\max}(z)&\equiv[\rho_{\max}^2+(z-z_i)^2]^{1/2}\ .\end{aligned}\tag{A6}$$

Now assume $kd\to0,k\epsilon\to0,d/\epsilon=\text{const}$. Then

$$\Gamma=-\frac{3}{2}\frac{\pi\beta n}{k^3}\left[2i+i\int_{z_i+\epsilon}^{d/2}\frac{dz}{(z-z_i)}+i\int_{-d/2}^{z_i-\epsilon}\frac{dz}{(z_i-z)}\right]=-\frac{3i\beta n\lambda^3}{8\pi^2}\left[1+\ln\left[\frac{(d^2/4-z_i^2)^{1/2}}{\epsilon}\right]\right]\ ,\tag{A7}$$

for $-d/2+\epsilon<z_i<d/2-\epsilon$. For $|z_i|=d/2$ we get by a similar calculation, the result

$$\Gamma=-\frac{3i\beta n\lambda^3}{16\pi^2}(1+\ln d/\epsilon)\ . \tag{A8}$$

APPENDIX B: CALCULATION OF Γ FOR A SPHERICAL SAMPLE

Let $\mathbf{r}_i=0$ and use spherical coordinates. Then, with $\mathbf{k}_0=\mathbf{k}\equiv k\hat{\mathbf{z}}$, and by using the approximation of Eq. (13), we have

$$\begin{aligned}\Gamma_1\equiv\sum_j{}' B(i,j)e^{ikz_j}&=\tfrac{3}{2}\beta n\int_0^{2\pi}d\phi\int_0^{\pi}d\theta\int_{r_{\min}}^{r_{\max}}dr\, r^2\sin\theta e^{ikr\cos\theta}[(\sin^2\theta\sin^2\phi+\cos^2\theta)F_{\rm I}(kr)+\sin^2\theta\cos^2\phi F_{\rm II}(kr)]\\&=\tfrac{3}{2}\pi\beta n\int_0^{\pi}d\theta\sin\theta\int_{r_{\min}}^{r_{\max}}dr\, r^2\sin^2\theta\, e^{ikr\cos\theta}[(\sin^2\theta+2\cos^2\theta)F_{\rm I}(kr)+\sin^2\theta F_{\rm II}(kr)]\ .\end{aligned} \tag{B1}$$

Let $x\equiv\cos\theta$; then,

$$\begin{aligned}\Gamma_1&=\tfrac{3}{2}\pi\beta n\int_{r_{\min}}^{r_{\max}}dr\int_{-1}^{1}dx\, r^2\cos(krx)\{F_{\rm I}(kr)+F_{\rm II}(kr)+x^2[F_{\rm I}(kr)-F_{\rm II}(kr)]\}\\&=3\pi\beta n\int_{r_{\min}}^{r_{\max}}dr\, r^2\left[(\sin kr)/kr[F_{\rm I}(kr)+F_{\rm II}(kr)]+\left[\frac{2\cos kr}{k^2r^2}+\frac{k^2r^2-2}{k^3r^3}\sin kr\right][F_{\rm I}(kr)-F_{\rm II}(kr)]\right]\end{aligned} \tag{B2}$$

or, with $kr=R$,

$$\begin{aligned}\Gamma_1&=\frac{6\pi\beta n}{k^3}\int_{R_{\min}}^{R_{\max}}dR\{\sin R\,[1/R-3/R^3-i(1-2/R^2+3/R^4)]+\cos R\,[3/R^2-i(1/R-3/R^3)]\}e^{iR}\\&=\frac{3\pi\beta n}{k^3}\int_{R_{\min}}^{R_{\max}}dR\{1+1/R^2+3/R^4+(2/R-6/R^3)\sin(2R)-(1-5/R^2+3/R^4)\cos(2R)\\&\qquad -i[(2/R-6/R^3)\cos(2R)+(1-5/R^2+3/R^4)\sin(2R)]\}\ .\end{aligned} \tag{B3}$$

Since the integrand is well behaved for small R [it is $O(R^2)+iO(R)$], we take $R_{\min}=0$ and let $R_{\max}\to R$. The integration then gives

$$\begin{aligned}\Gamma_1=\frac{3\beta n\lambda^3}{8\pi^2}&\left[R-\tfrac{1}{2}\sin(2R)-\frac{1+\cos(2R)}{R}+\frac{2\sin 2R}{R^2}-\frac{1-\cos(2R)}{R^3}\right.\\&\left.+i\left[-\tfrac{7}{6}+\tfrac{1}{2}\cos(2R)-\frac{\sin(2R)}{R}-\frac{2\cos(2R)}{R^2}+\frac{\sin(2R)}{R^3}\right]\right]\ .\end{aligned} \tag{B4}$$

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