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Effect of dipole-dipole interactions on optical bistability

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The effect of resonant collisions resulting from (Coulomb) dipole-dipole interaction is examined on optical bistability in the mean-field approximation. A propagator formulation is employed. A generalization of the standard equations of state for the absorptive-dispersive bistability is obtained. It is found that Lorentz-Lorenz local-field correction produces a small but appreciable shift of the optical-phase-transition point.

Optical bistability has received considerable attention in recent years.[1-3] In gases, the simple collisional aspects of the problem can be treated by using the transverse and longitudinal relaxation rates,[1,3] which is adequate for foreign gas and natural broadening. In this Brief Report we examine the modification of the standard results due to resonance broadening and Lorentz-Lorenz local-field correction, both of which arise from (Coulomb) dipole-dipole interaction.[4,5] The propagator formulation developed earlier[6] will be used for this purpose. (This paper will henceforth be referred to as I.) A simple approximation to the propagator equations leads directly to the mean-field theory. An equation of state is obtained which is valid for absorptive as well as dispersive bistability in a ring cavity. This equation is a simple generalization of the standard results, and reduces to these results if Lorentz-Lorenz correction is neglected. The effect of Lorentz-Lorenz correction on the phase transition is small, but not completely negligible (see Fig. 1).

We will consider the standard ring cavity[1] with the gas contained in the region, $0 \leq z \leq L$ (along z axis), and with end mirrors of reflection coefficient R. The absorbing atoms have a lower (upper) state $|a>$ ($|b>$). (In the resonance broadening limit the gas consists of identical atoms, but it could also include a buffer gas in the foreign broadening case.) Equations (2.5)–(2.9) of I can be easily modified to take account of the boundary conditions for this case. Combining Eqs. (2.5) and (2.9) of I and reducing the result to one-dimensional propagation (along the z axis with $\vec{k}=k\vec{e}_z$) inside the absorption cell, we obtain ($\hbar=1$)

$$\Omega(k_1,k,\omega)=\Omega'(k_1k\omega)+\int_{-\infty}^{+\infty}\frac{dk_2}{2\pi}\Gamma_0(k_1,k_2,\omega)\bar{\phi}(k_2,\omega)\,\Omega(k_2,k,\omega)\ , \tag{1}$$

$$\Omega'(k_1,k,\omega)=\sqrt{T}\,\Omega_0\Delta(k_1-k)+R\exp(i\theta)\,\Omega(k_1,k,\omega)\ , \tag{2}$$

$$\Gamma_0(k_1,k_2,\omega)=-4\pi\omega^2\int_{-\infty}^{+\infty}\frac{dk_3}{2\pi}\Delta(k_1-k_3)\frac{|d_{ab}|^2-|d_{ab}^3|^2}{\omega^2-c^2k_3^2+i0^+}\Delta(k_3-k_2)\ , \tag{3}$$

$$\Delta(k'-k)=\int_0^L dz\,e^{-i(k'-k)z}\ , \tag{4}$$

where Ω_0 is the Rabi frequency at resonance for the incident field, d_{ab}^3 is the component of the dipole matrix element $\vec{d}_{ab}$, along $\vec{k}_3=k_3\vec{e}_z$, and $T=1-R$, is the transmission coefficient of the end mirrors. Note that (2) simply represents the input boundary condition, at $z=0$, where θ is the mistuning with the nearest cavity mode. $\bar{\phi}$ is related to the dielectric susceptibility [see (8)].

To solve (1) in the mean-field approximation, we first obtain the appropriate approximation for Γ_0 from (3); evaluating the integral at the pole, $k_3=(\omega/c)+i0^+=k+i0^+$, we obtain

$$\Gamma_0(k_1,k_2,\omega)\approx i(2\pi/3)k|d_{ab}|^2\bar{\phi}(k,\omega)\Delta(k_1-k)\Delta(k-k_2)\ . \tag{5}$$

Here we have averaged over the dipole orientations so that $|d_{ab}^3|^2\to|d_{ab}|^2/3$. Now we use the large sample limit ($kL>>1$) in (4) to obtain

$$\Delta(k-k')\approx 2\pi\delta(k'-k)=L\delta_{k',k}\ . \tag{6}$$

Approximations (5) and (6) reduce (1) to an algebraic equation with the following solution:

$$\Omega(k)=\int_{-\infty}^{+\infty}\frac{dk_1}{2\pi}\Omega(k_1,k,\omega)\approx\frac{\Omega_0/\sqrt{T}}{1-i(\phi+2\pi kL\chi/T)}\ , \tag{7}$$

$$\chi(k)=\tfrac{1}{3}|d_{ab}|^2\bar{\phi}(k,\omega);\ \ \phi=\theta R/T\ . \tag{8}$$

Here it is assumed that $\theta<<1$, and χ denotes the dielectric susceptibility of the gas.[6] The above equation is a generalization of the standard equation of state in the mean-field theory.[1] First let us show that (7) reduces to the standard equation if Doppler effect and Lorentz-Lorenz correction are neglected, and the standard collision model is used. In this case,

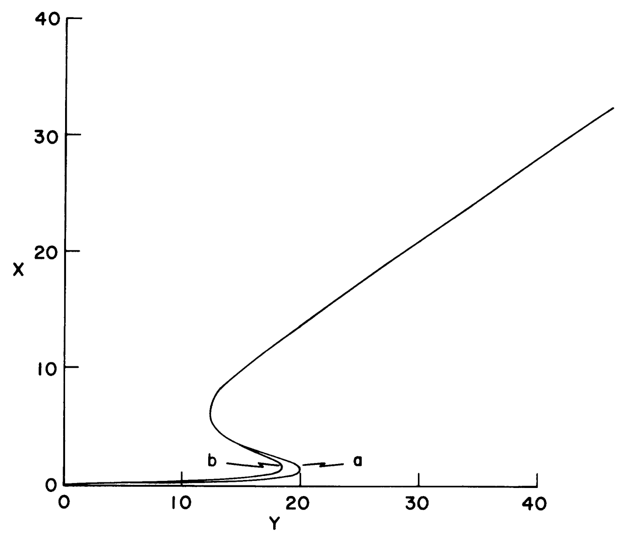

FIG. 1. Results predicted by the equation of state (15) are shown for two limiting cases. Here $C=20$, $\Delta=1$, $\phi=1$. Curve a: $Z=0$ (foreign gas and natural broadening limit); curve b: $Z=\frac{1}{3}$ (resonance broadening limit).

$$\bar{\phi}(\omega)=-n(\Delta\omega-i\gamma_\perp)(\Delta\omega^2+\gamma_\perp^2+\gamma_\perp\Omega^2/\gamma_\parallel)^{-1}\ ;\quad \Delta\omega=\omega-\omega_{ab}\ , \tag{9}$$

where $\gamma_\parallel$ ($\gamma_\perp$) is the longitudinal (transverse) relaxation rate, n is the number density of atoms, and $\Delta\omega$ is the detuning. For absorptive bistability, $\Delta\omega=0$, $\theta=0$, and (7)–(9) give

$$Y=X+2CX(1+X^2)^{-1}\ , \tag{10}$$

$$X=\Omega(\gamma_\perp\gamma_\parallel)^{-1/2};\quad Y=\Omega_0(\gamma_\perp\gamma_\parallel T)^{-1/2}\ , \tag{11}$$

$$C=\pi n|d_{ab}|^2kL(3\gamma_\perp T)^{-1}\ . \tag{12}$$

Equation (10) is the well-known equation of state for absorptive bistability.[1-3]

In this paper we will consider only the binary collisions in the impact limit.[7] Therefore many complications such as duration of collision effects,[4] many-body effects,[5] and the effect of the field on the collision rates[8] will be neglected. In this model, the resonance broadening arising from resonant collisions contributes to $\gamma_\perp$ in the usual manner. However, the contribution of the collisions in which excitation is transferred coherently from one excited atom to an unexcited one, through (Coulomb) dipole-dipole interaction, is taken into account in a generalized Lorentz-Lorenz local-field correction.[4] This correction is determined by T^e, the excitation transfer T-matrix element, according to (2.11) of I. The static limit value

$$T^e=-4\pi|d_{ab}|^2/9 \tag{13}$$

reproduces the standard value for Lorentz-Lorenz correction[4]; the motion and the spatial degeneracy can modify the multiplicative numerical factor.

Neglecting the energy and momentum dependence of T^e, Eqs. (2.10)–(2.14) of I lead to the following generalization of (9):

$$\bar{\phi}(\omega)=\frac{-n(\Delta\omega-i\gamma_\perp)}{\Delta\omega^2+\gamma_\perp^2+\gamma_\perp\Omega^2/\gamma_\parallel+nT^e(\Delta\omega-i\gamma_\perp)}\ . \tag{14}$$

Then (7), (8), and (14) give the following generalization of (10):

$$Y^2=X^2\left[\left(1+\frac{2C(1+X^2+\Delta^2)}{(1+X^2+\Delta^2+Z\Delta)^2+Z^2}\right)^2+\left(\frac{2C\Delta(1+X^2+\Delta^2+Z\Delta)+2CZ}{(1+X^2+\Delta^2+Z\Delta)^2+Z^2}-\phi\right)^2\right]\ , \tag{15}$$

$$\Delta=(\Delta\omega)/\gamma_\perp;\quad X=|\Omega|(\gamma_\parallel\gamma_\perp)^{-1/2};\quad Z=nT_e/\gamma_\perp\ . \tag{16}$$

Equation (15) reduces to the standard results if $Z\rightarrow 0$. In particular, it reduces to (10) if $\Delta=\phi=Z=0$. It follows that a similar equation can be obtained for a Fabry-Perot cavity if slightly different definitions of X, Y, and C are used. [See, e.g., Eq. (3.12) of Ref. 9 for the appropriate result for a Fabry-Perot cavity without Lorentz-Lorenz correction.]

Equation (15) is applicable to resonance broadening, foreign gas broadening, and natural broadening, if proper contributions to $\gamma_\parallel$ and $\gamma_\perp$ are included. For example, let $\gamma_\perp=\gamma_1+\gamma_2$, where γ_1 (γ_2) denote the resonance (foreign gas and natural) broadening rates. According to the simple collision model,[4] $\gamma_1\approx\pi^2n|d_{ab}|^2/3$. Then it follows from (13) and (16) that $Z\sim 4/3\pi\sim\frac{1}{3}$, in the extreme resonance broadening limit ($\gamma_2<<\gamma_1$). On the other hand, $Z\rightarrow 0$ in the foreign gas (and natural) broadening limit ($\gamma_2>>\gamma_1$). Therefore the effect of Lorentz-Lorenz correction is not expected to be very large. This effect is more clearly observable in the (mixed) absorptive-dispersive case. Figure 1 shows a comparison of the resonance broadening limit ($Z=\frac{1}{3}$) with the foreign gas (and natural) broadening limit ($Z=0$). The effect of Lorentz-Lorenz correction is appreciable in the region of the optical phase transition.

The general formulation of I covers not only the mean field, but also the fluctuations. For example, the fluorescence spectra are given by Eqs. (3.1)–(3.11) of I. These equations can be easily solved in the same manner as (7). The general formulation also contains the complete propagation, absorption, and scattering effects, and it can be developed into a theory which goes beyond the mean-field approximation.[1,10]

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