Phase transition induced by a laser field in a system of two-level atoms

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A new type of phase transition is considered, namely a laser-induced ferroelectric transition due to saturation of two-level atoms.

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It was shown in[1–3] that a soft mode can appear in the spectrum of the collective excitations of an equilibrium system of two-level atoms interacting via an electromagnetic field. Consequently, at a critical temperature T_c, a phase transition is possible such that at $T<T_c$ spontaneous atomic polarization sets in at zero frequency. T_c is determined from the equation[1,2]

$$D_c^{(\circ)} = \mathrm{th}\frac{\hbar\omega_{ab}}{2\kappa T_c} = \frac{1}{\beta\lambda_{ab}}; \qquad \lambda_{ab} = \frac{8\pi n\,|\mathbf{d}_{ab}\,\mathbf{e}_{\epsilon\circ}|^2}{\hbar\omega_{ab}}, \tag{1}$$

where $D_c^{(0)}$ is the difference between the populations of the levels a and b at $T=T_c$, ω_{ab} is the transition frequency, $\mathbf{d}_{ab}$ is the dipole matrix element, $\mathbf{e}_{\epsilon 0}$ is unit vector along the applied-field, $\beta(1\geqslant\beta>0)$ is the constant of the applied field ($\beta=1$ for the Dicke model[3] and $\beta=1/3$ for the Lorentz field), and n is the concentration of the atoms.

The induced phase transition considered here is possible in a non-centrosymmetric medium in which the working-level diagonal matrix elements $\mathbf{d}_{aa}$ and $\mathbf{d}_{bb}$ are different from zero. This transition takes place at realistic concentrations of the two-level atoms at easily attainable values of the laser intensity.

It is important that the induced phase transition is possible both in an equilibrium medium ($D<0$) and in a non-equilibrium inverted medium $D>0$. To this end the laser-emission frequency ω_L should be smaller than ω_{ab} in the first case (detuning $\Delta=\omega_{ab}-\omega_L>0$), and $\Delta<0$ in the second case.

For the population difference $D=\rho_{aa}-\rho_{bb}$ and for the function ρ_{ab} and $\rho_{ba}=\rho^*_{ab}$, we use the equations ($\rho_{aa}+\rho_{bb}=1$):

$$\frac{\partial}{\partial t} D + \gamma(D - D^{(o)}) = \frac{2i}{\hbar} (\mathbf{d}_{ab}\rho_{ba} - \rho_{ab}\mathbf{d}_{ba})(\vec{\epsilon} + \mathbf{E}_L), \tag{2}$$

$$\left(\frac{\partial}{\partial t} + i\omega_{ab} + \gamma_{ab}\right)\rho_{ab} = -\frac{i\mathbf{d}_{ab}}{\hbar} D(\vec{\epsilon} + \mathbf{E}_L) + \frac{i}{\hbar}\mathbf{d}\rho_{ab}(\vec{\epsilon} + \mathbf{E}_L). \tag{3}$$

Here $\mathbf{d}=\mathbf{d}_{aa}-\mathbf{d}_{bb}$, $\vec{\epsilon}=\mathbf{e}_\epsilon=\mathbf{E}+\beta 4\pi\mathbf{p}$ is the effective field, $\mathbf{E}$ is the intrinsic field, $\mathbf{E}_L$ is the given laser field

$$\mathbf{E}_L = \mathbf{e}_L(E_L e^{-i(\omega_L t - K_L R)} + \text{c.c.}) \tag{4}$$

and $D^{(0)}$ is a given quantity, in particular, the equilibrium population difference.

From Maxwell's equations for $\mathbf{E}$ we obtain for the field $\vec{\epsilon}$ the equation

$$\frac{\partial^2\vec{\epsilon}}{\partial t^2} - c^2\Delta\vec{\epsilon} = (\beta-1)4\pi\frac{\partial^2\mathbf{p}}{\partial t^2} - \beta 4\pi c^2\Delta\mathbf{p}. \tag{5}$$

The polarization vector $\mathbf{p}$ at $\mathbf{d}\neq 0$ can be represented in the form

$$\mathbf{p} = \mathbf{p}_{ab} + \mathbf{p}_D; \qquad \mathbf{p}_{ab} = n(\mathbf{d}_{ab}\rho_{ba} + \mathbf{d}_{ba}\rho_{ab}), \quad \mathbf{p}_D = \frac{n}{2}\mathbf{d}D. \tag{6}$$

The effective field $\vec{\epsilon}$ has a fast component ($\omega\sim\omega_{ab}$) and slow component ($\omega\ll\omega_{ab}$). Under the condition $|E_L|\gg|\epsilon(\omega)_{ab}|$, Eqs. (2) and (3) yield

$$D = D^{(o)}\frac{\left[\left(\Delta - \frac{\mathbf{d}\epsilon_o}{\hbar}\right)^2 + \gamma_{ab}^2\right]}{\left(\Delta - \frac{\mathbf{d}\vec{\epsilon}_o}{\hbar}\right)^2 + \gamma_E^2}; \qquad \begin{aligned} a &= 4|\mathbf{d}_{ab}\mathbf{e}_L|^2/\hbar\gamma\gamma_{ab} \\ &= \gamma_{ab}^2(1 + a|E_L|^2) \end{aligned}, \tag{7}$$

where ϵ_0 is the slowly varying part of the effective field.

We see that when the laser radiation is applied the average difference of the atom-level populations, and hence the average value of its dipole moment $\mathbf{d}D$, depends not only on $a|E_L|^2$, but also on the relative orientation of the dipole-moment vector $\mathbf{d}$ and the vector $\vec{\epsilon}_0$. It is this which makes posssible, as we shall show, the induced phase transition.

In fact, from (6) and (7) we get in the linear approximation in ϵ_0

$$4\pi(\mathbf{e}_{\epsilon_0}\mathbf{p}_D) = -\lambda_L D_0 \epsilon_0;\ \lambda_L = \frac{4\pi(\mathbf{d}\mathbf{e}_{\epsilon_0})^2 n}{\hbar\omega_{ab}} \frac{\Delta\gamma_{ab}^2\omega_{ab}}{(\Delta^2+\gamma_E^2)(\Delta^2+\gamma_{ab}^2)} a|E_L|^2, \quad (8)$$

where D_0 is determined by expression (7) in the zeroth approximation in ϵ_0. Substituting (8) in (5) and using also the equation that follows from (3) for $\mathbf{p}_{ab}$, we get the spectrum of the collective excitations $\omega=\omega(\mathbf{k})$, where $\mathbf{k}$ is the wave vector. If the condition

$$1+\beta(\lambda_{ab}+\lambda_L)D_0 = 0 \quad (9)$$

is satisfied, then $\omega(\mathbf{k})$ vanishes at all values of $\mathbf{k}$.

If there is no laser radiation and the system is in equilibrium, then condition (9) coincides with (1). At $|\Delta|\sim\gamma_{ab}$, $a|E_L|^2\sim 1$ and $|\mathbf{d}|\sim|\mathbf{d}_{ab}|$ we have the ratio $|\lambda_L|/\lambda_{ab}\sim\omega_{ab}/\gamma_{ab}\gg 1$. Thus, the induced contribution is dominant under these conditions.

To find the order parameter for the induced phase transition, we use Eq. (5), where $\mathbf{p}_D$ is determined by the expansion of (7) in powers of the parameter $\mathbf{d}\epsilon_0/\hbar\Delta$ (the contribution of $\mathbf{p}_{ab}$ can be neglected if $|\lambda_L|\gg\lambda_{ab}$). After averaging over the directions of the vector $\mathbf{d}$, we obtained an equation for the order parameter in the standard form of the Landau theory of phase transitions

$$a\epsilon_0 + b\epsilon_0^3 + c\epsilon_0^5 = 0, \quad (10)$$

where the coefficients are

$$a = 1+\beta\lambda_L D_0 \qquad (\lambda_L D_0 < 0),$$

$$b = \beta\lambda_L D_0 \frac{2(\Delta^2-\gamma_E^2)}{(\Delta^2+\gamma_E^2)^2}\left(\frac{\mathbf{d}\mathbf{e}_{\epsilon_0}}{\hbar}\right)^2,$$

$$c = \beta\lambda_L D_0 \frac{3(\gamma_E^4+\Delta^4)-10\gamma_E^2\Delta^2}{(\Delta^2+\gamma_E^2)^4}\left(\frac{\mathbf{d}\mathbf{e}_{\epsilon_0}}{\hbar}\right)^4. \quad (11)$$

At $\Delta^2<\gamma_E^2$, a second-order transition takes place. The fifth-power term can be neglected ($c=0$) and the order parameter is $\epsilon_0=\pm\sqrt{|a|/b}$. At $\Delta^2=\gamma_E^2$, we have $b=0$

and it follows from (11) that $c=\frac{1}{4}\beta|\lambda_L D_0|(\mathbf{d}\cdot\mathbf{e}_{\epsilon_0}/\hbar\gamma_E)^4$ and the order parameter is $\epsilon_0=(|a|/c)^{1/4}$. At $\Delta^2>\gamma_E^2$ the coefficient b becomes negative, but $c>0$ and a first-order transition takes place.

We have considered a nonequilibrium phase transition induced by an external field. It is of interest to consider an induced phase transition in the laser working medium. In this case the field causing the phase transition is generated in the same substance and consequently the lasing and the self-induced phase transition can effect each other. We note, in particular, the possibility of periodic breaks in the lasing (a spike regime) because of the variation of the frequency of the working transition $\omega_{ab}(E_L)=\omega_{ab}-\mathbf{d}\epsilon_0/\hbar$ [see (3)] when a static field $\vec{\epsilon}_0=\vec{\epsilon}_0(E_L)$ appears.

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