LETTER

Dynamical instabilities of spectroscopic transitions in dense resonant media

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Abstract

We consider the influence of the near dipole–dipole interaction, underlying the local field, on the dynamics of two-level and V-type three-level atoms exposed to cw-laser radiation. The near dipole–dipole interaction between the V-type three-level atoms is shown to give rise to the Poincaré–Andronov–Hopf bifurcation in steady-state solution of the equations of motion. As a result, the populations of the energy levels periodically vary in time. In the framework of the nondegenerate two-level model, dynamical instabilities are proved to be impossible.

(Some figures may appear in colour only in the online journal)

1. Introduction

The behavior of a single resonant atom in an external electromagnetic field is crucially dependent on the presence of other atoms of the same or other type. As a rule, if the density of resonant atoms in a medium is sufficiently low a single resonant atom is assumed to interact with the macroscopic field averaged over all the medium. By contrast, in optically dense media, where the density of resonant atoms is high enough, resonant atoms cannot be considered as independent ones and the theoretical models should be corrected for the atom–atom interaction. This interaction may include a number of processes of different nature and leads to complex response of media to an external field. One such process is the near dipole–dipole (NDD) interaction which becomes more conspicuous with increasing density of resonant atoms, and the behavior of a single resonant atom is influenced by the environment via field-induced dipoles.

Following [1], allowance for the NDD interaction is made by substituting the local field correction (LFC) into the Bloch equations (or the density matrix equation). Formally, LFC consists of the replacement of the macroscopic field $E$ in the Bloch equation by the local (or effective) one $E_{loc}$, according to the relation $E_{loc} = E + 4\pi P/3$, where $P$ is the induced macroscopic polarization. This phenomenological procedure of introducing LFC is equivalent to a consistent derivation of the Bloch equations from the Bogoliubov–Born–Green–Kirkwood–Yvon hierarchy for the reduced single particle density matrices of atoms and quantized field modes [2].

The LFC causes nonlinearity in the Bloch equations, and it is precisely this feature that has enabled a number of phenomena to be predicted: the intrinsic optical bistability (IOB) [3, 4]; Lorentz shift [5, 6] and anomalous excitation dependence of spectral line broadening [7, 8]; ultrafast intrinsic optical switching [9]; enhancement of the absorptionless index of refraction and inversionless gain in dense, coherently prepared, three-level atoms [10, 11]; enhancement of the spontaneous decay rate [12]; modification of the superradiant amplification characteristics [13]; incoherent soliton formation and phase modulation emergence in self-induced transparency [14]; shifts of the Autler–Townes peaks in absorption spectra [15]; optical switching operation, stable and unstable hysteresis...
loops, and an auto-oscillation regime of reflectivity [16, 17]. The influence of NDD interaction on pattern formation in broad-area cavity lasers and reduction of the second (chaotic) laser threshold in single-mode homogeneously broadened lasers is given in the review [18].

Among the most dramatic examples of the NDD effect, dynamical oscillations appear to be of particular interest in theoretical and practical terms. The present report is just devoted to this problem. More specifically, our principal concern is with dynamical instabilities developing in the dynamics of purely spectroscopic transitions, leaving aside propagation, boundary and retardation effects as well as laser regimes; this offers a fresh approach to the problem we have touched on.

The idea of dynamical instability due to the NDD interaction was originated from the dynamics of nonlinear systems. From the formal point of view of nonlinear dynamics, the generalized Bloch equations (even for a two-level atom) can produce a non-steady response to an external electromagnetic field with a constant intensity. However, in the literature the complete stability analysis is often omitted [10, 11, 19] or restricted to the existence of IOB [3, 4], assuming the steady-state response of a dense medium only. Disregarding the possibility of other kinds of instability in dense media can lead to limitation of the results obtained earlier and incorrect theoretical predictions. The nature of such an instability (discussed below), unlike the case in [16, 17], is not related to the saturation and propagation effects or local IOB and manifests itself independently.

To put it differently, the motivation of the research here would be expressible as a question: where and how (or under what conditions) could the NDD interaction result in dynamical instabilities when dealing only with spectroscopic transitions?

The search for relevant conditions, if any, is supposed to answer the question posed. For this purpose we consider a thin layer of optically dense medium where the resonant atoms are modeled by two- and three-level (V-type) quantum systems. Such a model enables us to neglect the retardation effects and the far-zone part of the local field generated by the dipoles as well as collective radiative relaxation processes (see [3, 20]). Based on these assumptions, we analytically prove the impossibility of the dynamical instabilities in the framework of the nondegenerate two-level atom model and show numerically that the instability is realizable in a certain range of the parameters of the three-level V-type atom model and discuss the experimental conditions for observation of the effect.

It is worth noting that, in the case of optically dense V-type media, the first step in studies of the NDD effect seemed to be taken by the authors of [19]. However, we have taken a further look at the properties of the V-system influenced by the NDD interaction to better appreciate the significance of the NDD effects while arriving at instabilities.
where $\Delta_{21} = \omega_{21} - \omega_0$ is the detuning of the resonant transition frequency $\omega_{21}$ from the electromagnetic field frequency $\omega_0$. $\Gamma_\parallel \equiv \Gamma_{12} = 4 |\mu_{21}|^2 k_A^2 l_3 (n) / (3h)$ is the relaxation constant (spontaneous emission rate), $h$ is the Planck constant, $k_A = \omega_{21} / c$, $c$ is the velocity of light in vacuum, $\Omega_{12} = \Omega_{12}^2 = \mu_{21} E_0 l_2 (n) / h$ is the Rabi frequency, $E_0$ is the slowly varying amplitude of the electromagnetic field $E = E_0 e^{-i\omega t} + c.c.$, $\mu_{21}$ is the transition dipole moment, $a = 4 \pi n_A |\mu_{21}|^2 l_3 (n) / (3h)$ is the LFC parameter (Lorentz frequency shift), and $n_A$ is the density of atoms. The correction factors $l_3(n)$ take into account the influence of a dielectric medium (host) with the refractive index $n$ and depend on the impurity type [2]. $\gamma = \Gamma_\parallel / \Gamma_{12}$ is the dephasing rate constant in units of the population relaxation rate constant ($\gamma = 1/\tau$ for the natural linewidth broadening only).

On substitution of operators (2) into (1) and transformation of the complex equations to real ones, the nonlinear system of Bloch equations reads

$$ V_A = -\begin{pmatrix} 0 & \Omega_{12} \\ \Omega_{21} & 0 \end{pmatrix}, \quad V_C = -\omega_{21} \begin{pmatrix} 0 & \rho_{12} \\ \rho_{21} & 0 \end{pmatrix}, \quad H_0 = \begin{pmatrix} 0 & 0 \\ 0 & \Delta_{21} \end{pmatrix}. \quad (2) $$

where $\Delta_{21} = \omega_{21} - \omega_0$ is the detuning of the resonant transition frequency $\omega_{21}$ from the electromagnetic field frequency $\omega_0$. $\Gamma_\parallel \equiv \Gamma_{12} = 4 |\mu_{21}|^2 k_A^2 l_3 (n) / (3h)$ is the relaxation constant (spontaneous emission rate), $h$ is the Planck constant, $k_A = \omega_{21} / c$, $c$ is the velocity of light in vacuum, $\Omega_{12} = \Omega_{12}^2 = \mu_{21} E_0 l_2 (n) / h$ is the Rabi frequency, $E_0$ is the slowly varying amplitude of the electromagnetic field $E = E_0 e^{-i\omega t} + c.c.$, $\mu_{21}$ is the transition dipole moment, $a = 4 \pi n_A |\mu_{21}|^2 l_3 (n) / (3h)$ is the LFC parameter (Lorentz frequency shift), and $n_A$ is the density of atoms. The correction factors $l_3(n)$ take into account the influence of a dielectric medium (host) with the refractive index $n$ and depend on the impurity type [2]. $\gamma = \Gamma_\parallel / \Gamma_{12}$ is the dephasing rate constant in units of the population relaxation rate constant ($\gamma = 1/\tau$ for the natural linewidth broadening only).

To perform the stability analysis of the steady-state solutions $\bar{r}_{ij}$ (4), it is necessary to linearize the system in the vicinity of $\bar{r}_{ij}$, construct the Jacobi matrix $J(\bar{r}_{ij})$ and examine its eigenvalues. Then the eigenvalue problem is reduced to the solution of the characteristic polynomial with respect to the eigenvalues $\lambda_i$:

$$ \det (J(\bar{r}_{ij}) - \lambda I) = a_3 \lambda^3 + a_2 \lambda^2 + a_1 \lambda + a_0 = 0. \quad (5) $$

where $I$ is the $3 \times 3$ identity matrix, the coefficients $a_i$ being cumbersome expressions that are not represented here.

In the search for instabilities, there is a need to find conditions when the real parts of eigenvalues $\lambda_i$ become positive. So, when the complex eigenvalue intersects the imaginary axis of the complex plane, we can represent the eigenvalues as $\lambda = \rho + i\xi$, where $\xi$ is real, and substitute them into (5). Upon separating the resulting equation into real and imaginary parts and eliminating $\xi$, we get the conditions whereby the real part of the eigenvalue changes the sign:

$$ a_0 = 0 \quad \text{or} \quad a_0 a_{31} - a_1 a_{21} = 0. \quad (6) $$

It follows from equation (5) that the first condition of (6) corresponds to the sign change of the real eigenvalue of the Jacobi matrix and leads to the IOB occurrence. In the explicit form it can be expressed as

$$ \gamma^2 + (\beta - \delta)^2 - 4\beta r_{22} (2(\beta - \delta) \quad + r_{22}(-5\beta + 2\delta + 4\beta r_{22})) = 0, \quad (7) $$

where we formally regard a steady value of the population $r_{22}$ of the upper level as an independent variable in the interval $0 \leq r_{22} < 0.5$ (the condition of impossibility of population inversion in the framework of the two-level atom model). The relation between $r_{22}$ and the dimensionless amplitude of the Rabi frequency $\Phi$ is given by steady-state solutions (see the last equation (4)). Roots of equation (7) define the critical points of a hysteresis loop of IOB as well as a condition of its occurrence (it turns out to be $(\beta - \delta)^2(\beta + 8\delta) > 27\gamma^2\beta$).

The explicit form of the second condition (6) can be written as

$$ (1 - 2r_{22})(r_{22} \gamma + (1 - 2r_{22})\gamma^2)(\gamma(1 + \gamma) \quad + (\delta - (1 - 2r_{22})\beta^2) + (1 - 2r_{22})(\gamma^2(1 + \gamma) \quad - r_{22}\beta\delta) + r_{22}\delta^2) = 0. \quad (8) $$

Equation (8) has real roots and predicts the Poincaré–Andronov–Hopf bifurcation occurrence (the emergence of a pair of complex eigenvalues with positive real parts) only outside the interval of physically allowed values of the upper level population $0 \leq r_{22} < 0.5$ and at $\Phi^2 < 0$. This implies the absence of dynamical instabilities in the framework of the two-level atom model without the degeneracy.

In spite of this, there is a hope to find oscillations (or chaos) in more complex cases of configurations of the atomic energy levels. For our analysis, among all the possible configurations of energy levels of three-level atoms, we choose the V-type level system and perform analysis numerically because of complexity associated with a great number of dynamical variables and parameters.

### 2.2. Dense media of three-level V-type atoms

Let us assume that the three energy levels of a single atom have the V-type configuration with the ground state $|1\rangle$ and two excited states $|2\rangle$ and $|3\rangle$ (see figure 1(b)). The transition $|2\rangle \leftrightarrow |3\rangle$ is forbidden (the transition dipole moment $\mu_{23} = 0$) and the remaining transitions $|1\rangle \leftrightarrow |2\rangle, |3\rangle$ are coupled by one and the same laser field. The transition frequencies
obey the condition $\omega_{32} \ll \omega_{21}, \omega_{31}$. The absolute values of the dipole transition dipole moments are $\mu = |\mu_{21}| = |\mu_{31}|$ and orthogonal; that is, $(\mu_{21}, \mu_{31}) = 0$ (the quantum interference is absent). It is also anticipated that the stationary electromagnetic field has the frequency $\omega_{0}$ and the detunings from the resonant transitions' frequencies $\omega_{21}$ and $\omega_{31}$ are equal $\Delta_{21} = \omega_{31} - \omega_{0}$. At the chosen parameters the Rabi frequencies, relaxation constants and local field parameters are $\Omega = \Omega_{12} = \Omega_{13}, \Gamma_{\parallel} = \Gamma_{12} = \Gamma_{13}, \omega_{L} = \omega_{L}^{21} = \omega_{L}^{31}$. In doing so, operators of (1) have the form

$H_{0} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & \Delta_{21} & 0 \\ 0 & 0 & \Delta_{31} \end{pmatrix}$, \hspace{1cm} V_A = -\begin{pmatrix} 0 & \Omega & \Omega \\ \Omega^* & 0 & 0 \\ \Omega^* & 0 & 0 \end{pmatrix}$, \hspace{1cm} \Gamma (\rho) = -\Gamma_{\parallel} \begin{pmatrix} -\rho_{22} - \rho_{33} & \gamma_{12}\rho_{12} & \gamma_{13}\rho_{13} \\ \gamma_{12}\rho_{12} & \rho_{22} & \gamma_{23}\rho_{23} \\ \gamma_{13}\rho_{13} & \gamma_{23}\rho_{32} & \rho_{33} \end{pmatrix}$, \hspace{1cm} (9) \hspace{1cm} V_C = -\omega_{L} \begin{pmatrix} 0 & \rho_{12} & \rho_{13} \\ \rho_{21} & 0 & 0 \\ \rho_{31} & 0 & 0 \end{pmatrix}$, \hspace{1cm} \gamma_{ij} = \frac{\Gamma_{\parallel}^{ij}}{\Gamma_{\parallel}}$ are the phenomenological dephasing rate constants in units of the population relaxation rate constant.

First, we consider the simplest case, where the line broadening is associated with the natural width only (the relaxation constant ratios yield $\gamma_{12} = \gamma_{31} = \gamma_{23}/2 = \frac{1}{2}$, and $\Delta = \Delta_{31} = -\Delta_{21}$. Substituting (9) into (1) gives the same complex set of differential equations as in [19], where use is made of the phenomenological approach ($E \rightarrow E_{loc}$) but with allowance for the quantum interference. For the numerical simulations we transform the obtained complex equations into the eight real ones (in a way analogous to (3)) to perform the numerical stability analysis of a steady-state solution and direct numerical integration of the obtained real nonlinear equations.

Figure 2 illustrates the steady-state solutions for the energy level population $\rho_{22}$ and $\rho_{33}$ versus the Rabi frequency $\Phi$ at $\delta_{31} = -\delta_{21} = \Delta_{31}/\Gamma_{\parallel} = -\Delta_{21}/\Gamma_{\parallel} = 1.75$, $\beta = \omega_{L}/\Gamma_{\parallel} = 6$, and appropriate solutions of original differential equations after the integration time $\tau = 100$. For comparison, the evolution of level populations at a negligibly small NDD interaction ($\beta = 0$) is shown by a thin solid line. Figure 2 also demonstrates the evolution of the real and imaginary parts of the complex eigenvalue obtained from the linear stability analysis of the steady-state solution. It is remarkable that there is an unstable region where there exists a pair of complex conjugated numbers with a positive real part. This is indicative of the Poincaré–Andronov–Hopf bifurcation occurrence, which predicts oscillations of the density matrix elements $\rho_{ij}$. Thus, we have answered the above-posed question. That is, the NDD interaction can lead...
Figure 3. Temporal dynamics of the population of energy levels $\rho_{22}$ (solid line) and $\rho_{33}$ (dashed line) after the transient-process time $\tau = 100$ in the instability region of figure 2 at $\Phi = 1.1$ (a) and the corresponding attractor projection (b) onto the plane $(\rho_{22}, \rho_{33})$.

Figure 4. Dynamical instability (a) in (1) with operators (9) at the parameters $\delta_{21} = -2$, $\delta_{31} = 6$ and $\beta = 6$, which corresponds to $\delta = -2$ (dot-dashed line) in the stability map (b) in coordinates of $\delta = ((\omega_{31} + \omega_{21})/2 - \omega_{0})/\Gamma_{1}$ versus natural steady curve parameter $s$. The dashed lines in (a) correspond to the steady stable parts of the curve, black dots to the unstable one, red ($\rho_{22}$) and green ($\rho_{33}$) dots show the numerical solution of the original differential equation and a thin solid line indicates the appropriate evolution of the energy level population but without NDD interaction. Areas 1 and 2 in (b) are the bistability region and area 3 is the oscillation region. The thin solid line in (b) indicates the resonances with energy levels of V-type atoms and the dashed line corresponds to the Rabi frequency $8 = 20$ (where the calculation stops).

to the dynamical instability, and to this end V-system-based media are quite promising. The direct numerical integration of the system reveals the oscillations in the unstable region (see figure 2). The dynamics of the level population and projection of the corresponding attractor onto the plane $(\rho_{22}, \rho_{33})$ at the Rabi frequency $\Phi = 1.1$ are shown in figure 3. As seen from figure 3, the behavior of temporal changes in the populations has a periodic nature, the oscillations being displaced in phase. The calculation of the Lyapunov exponent spectrum according to algorithms presented in [21] (it turns out to be $\{0, -, -, -, -, -, -, -\}$) also confirms the periodicity of oscillations. In our consideration, the quantum interference is not taken into account; in accordance with the aforesaid, $(\mu_{21}, \mu_{31}) = 0$ and in this case the effect predicted is most pronounced; otherwise, oscillations are completely suppressed.

In figure 2 the oscillations are depicted against the background of IOB. Meanwhile, there are parameters for which the oscillations exist without the hysteresis loop. Figure 4(b) displays the stability map for parameters $\beta = 6$ and the existence of oscillations without IOB figure 4(a) at $\delta_{21} = -2$ and $\delta_{31} = 6$. In figure 4(b) the vertical axis represents the electromagnetic field frequency change over the region while the horizontal axis represents the natural parameter $s$ of a steady curve in the eight dimensional phase space. In real systems the linewidth can exceed the natural linewidth due to the interaction of atoms with the environment and result in homogeneous and inhomogeneous broadening. The influence of the homogeneous line broadening mechanism is shown in figure 5. As seen from the figure, the amplitudes of oscillations as well as their region are small.
Figure 5. The same as in figures 2(a) and (b) but for $\beta = 500$, $\delta = 55$, $\gamma_{12} = \gamma_{13} = 10$ and $\gamma_{23} = 4$. The parameters chosen are close to the oscillation threshold.

The increase in $\beta$ leads to the enhancement in amplitude and to the extension of the region of oscillations. According to calculations, the existence condition of the predicted effect (similar to the IOB threshold) requires that $\omega_L > \sum_{ij} \Gamma_{ij}^\perp$ for the absence of the pure dephasing. With increasing dephasing rate constant, the threshold condition of oscillations becomes $\omega_L \gg \sum_{ij} \Gamma_{ij}^\perp$ (compare the parameters in figures 2 and 5). Estimations show that the threshold conditions cannot be satisfied for $\gamma_{ij} > 10^2$. So we can conclude that the existence condition of population oscillations is the comparability (within one order) of the relaxation rate constants. From the subsequent analysis it is apparent that any additional relaxation process in a system (relaxation between excited levels, collective relaxation effects) will merely suppress oscillations as an additional ‘friction’ in a nonlinear system (this also follows from the basic principles of dynamics of nonlinear systems). The influence of the inhomogeneous broadening makes the problem much more complex, although some preliminary conclusions can be made. It is expected that the inhomogeneous broadening can give rise to a more complex dynamics of the system and produce the chaotic behavior instead of the periodical one as well as suppressing the effect, depending on the type of the inhomogeneous distribution function.

3. Estimation of experimental conditions

It remains for us to briefly discuss the feasibility of experimental observation of the predicted exhibition of the local field effect. The predicted effect can take place in crystals doped by rare earth ions at the concentration higher than 1 at.%. In addition to the increase in the concentration, the homogeneous linewidth should be reduced by an appropriate choice of host media and cooling them to temperatures below 10 K. This results in the comparable relaxation rates of population and polarization ($\Gamma_{\perp}/\Gamma_{\parallel} \approx 3$ for Pr$^{3+}$:Y$_2$SiO$_5$ [22]. Spectrally isolated three-level systems can be prepared with the help of spectral hole burning techniques [23]. Here, it is preferable to minimize the inhomogeneous broadening to increase the number of ions in spectral holes.

The possibility of creating quantum dot (QD) arrays with low inhomogeneity ($< 1$ meV) [24] and surface density up to $10^{11}$ cm$^{-2}$ [25] allows one to propose an alternative system to observe oscillations. The V-type three-level system can be realized by fine-structure states of a single QD with orthogonal transitions at low temperatures ($< 10$ K), where the linewidth is primarily lifetime limited with $\Gamma_{\perp} \sim 10^{10}$ Hz [26]. Single QD parameters of [27–29] were used to show the effect of population oscillations pictured in figure 2. The estimations show that the effect is observable at concentration higher than $10^{15}$ cm$^{-3}$. The technological tendency of creating QD arrays with a higher homogeneity ($< 100$ µeV), which is required for quantum computing [24], gives hope to attain a broadening ratio of the order of unity.

4. Conclusion

The main findings of our investigation into dynamical spectroscopic-transition instabilities in dense media are as follows. Oscillations associated with instabilities are in principle impossible in the most primitive ensembles of two-level non-degenerate atoms. Fundamentally, to induce these oscillations, what is wanted is a definite complexity of the level structure, namely, the number of levels must be greater than two. This is exemplified by the V-type three-level atom model. Just in this case the NDD interaction can lead to the dynamical instabilities via Poincaré–Andronov–Hopf bifurcation while solving the equations of motion. It is established that the instabilities can emerge in some range of the relationship between the rates of transversal and
longitudinal relaxations, which is quite achievable at low temperatures. The instabilities can develop regardless of IOB; in particular, they can occur against the background of IOB. In any case the oscillations exhibited are of an intrinsic nature peculiar to dense resonant media. In essence, a new type of dynamical instability (in dense media) is found, which is the first of its kind ever revealed. In order for our model to approach reality, we suggest some credible materials where oscillations can be brought about; their parameters are best suited to our purposes.

The obtained results open new vistas of optics of dense resonant media and it is obvious that the predicted effect can manifest itself in the case of more complex level configurations such as lambda, cascade and degenerate two-level systems. Undoubtedly, this is a new aspect of dynamical instability, which seems to deserve further investigations and should be taken into account in the theoretical analysis and interpretation of experimental results related to optically dense media.

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