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# Fast electromagnetic response of a thin film of resonant atoms with permanent dipole

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## Abstract

We consider the propagation of extremely short pulses through a dielectric thin film containing resonant atoms (two-level atoms) with permanent dipole. Assuming that the film width is less than the field wavelength, we can solve the wave equation and reduce the problem to a system of generalized Bloch equations describing the resonant atoms. We compute the stationary solutions for a constant irradiation of the film. Superimposing a small amplitude linear wave we calculate the reflection and transmission coefficients. From these, one can then deduce the different parameters of the model. We believe that this technique could be used in experiments to obtain the medium atomic and relaxation parameters.

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(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

One of the famous models describing field–matter interaction is the Maxwell–Bloch system [1]. It corresponds to an ensemble of two-level atoms whose states alternate due to the electromagnetic field. In a simple approach the electromagnetic field is assumed to be scalar and the operator of dipole transition to only have non-diagonal matrix elements. This model was the base for the description of many coherent nonlinear effects, the coherent pulse propagation (self-induced transparency [2]) and the coherent transient effects (optical nutations, free induction decay, quantum beats, superradiance, photon echo). A detailed review of these phenomena may be found in [3, 4] and in the book [1].

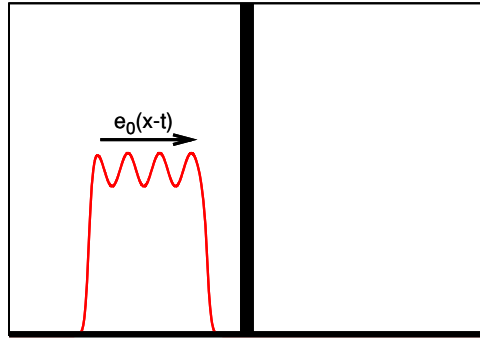
The Maxwell–Bloch system can be extended further than the model of two-level atoms discussed above. In particular one can generalize the resonant atomic model. The model of three-level atoms now attracts great interest, because it describes electromagnetic induced transparency, slow light propagation in (three-level) atomic vapor [5] and the coherent population transfer [6]. The coherent interaction of electromagnetic pulses with quantum dots [7, 8] can be described by such a generalized Maxwell–Bloch system. Another generalization of the two-level model is to take into account diagonal matrix elements of the dipole transition operator [12]. In this medium, steady state one-half cycle pulses were obtained in the sharp line limit [13]. A new kind of steady state pulse was found, characterized by an algebraic decay of the electric field.

The reduced Maxwell–Bloch equations in the sharp line limit have a zero-curvature representation [14, 15]. A number of results related to the complete integrability of this reduced system have been obtained in [16–18]. The numerical simulation of the propagation of extremely short pulses [19] shows the existence of an extraordinary breather with non-zero pulse square. An analytical expression for this breather has been found in [14, 15] and reproduced in [20] using the Darboux transformation method. Recently, Zabolotzki [21] developed the inverse scattering method to find the solution for the isotropic limit of the general model of a two-component electromagnetic field interacting with two-level atoms with a permanent dipole moment without invoking the slowly varying envelope approximation.

The influence of the permanent dipole on parametric processes was studied in [9, 10]. Recently, Weifeng *et al* studied the generation of attosecond pulses in a two-level system with permanent dipole moment [11]. In this case higher harmonics are generated and the spectrum can be extended to the x-ray range. The quantum interference of both even and odd harmonics results in the generation of higher intensity attosecond pulses.

The propagation of ultrashort pulses through a thin film containing resonant atoms located at the interface between two dielectrics is also described by the Maxwell–Bloch equations. However, if the width of the film is less than a wavelength [22], the atomic system is compressed into a ‘point’. An insightful comment was made in [23, 24], concerning the critical role of the local Lorentz field. This local field induces a nonlinearity so that the thin film of two-level atoms acts as a nonlinear Fabry–Perot resonator. One then expects optical bistability for this device. There are many generalizations of the thin film model, where three (or more) level atoms, two-photon transition between resonant levels and non-resonant nonlinearity were studied. Here we consider a thin film corresponding to two-level atoms with permanent dipole moment [25]. There the author analyzed numerically the pulse propagation through the film taking into account the local field. He showed that a dense film irradiated by a one-cycle pulse emits a short response with a delay much longer than the characteristic cooperative time of the atom ensemble. Contrary to [25] we assume that the atoms of the film are rare so that the local field can be neglected.

Recently [27], we introduced a general formalism to describe the interaction of a (linear polarized) electromagnetic pulse with a medium. For a thin film, the solution of the linear problem (the wave equation) can be folded back into the nonlinear problem. With this we studied a ferroelectric film which was described by a Duffing oscillator naturally giving a double-well potential. Here we generalize this approach to the case of a layer of resonant atoms described by the generalized Bloch equations taking into account the permanent dipole moment. As for the ferroelectric problem, the field is given in terms of the film variables. An important feature of the model is that there is a clear separation between the medium and the surrounding vacuum so that even in the linear case no dispersion relation can be written. Instead we obtain a scattering problem where the reflection and transmission coefficients need to be calculated.



**Figure 1.** Schematic of an incoming electromagnetic pulse  $e_0$  on a thin dielectric film containing resonant atoms.

After presenting the model in section 2, we compute its equilibria and their stability in section 3. In section 4 we assume an additional periodic modulation around a fixed background field. This enables us to do a spectroscopy study of the film so that both the dipole parameter and the coupling parameter can be extracted from the reflection curve. Section 5 concludes the paper.

## 2. The model

### 2.1. One-dimensional wave propagation

Following the formalism of [27] we assume that an electromagnetic wave is incident from the left  $x < 0$  on a medium whose position is given by the function  $I(x)$ . The configuration is shown in figure 1. Denoting by subscripts the partial derivatives, the equations are

$$e_{tt} - e_{xx} = g(x, t), \tag{1}$$

$$g(x, t) = -\gamma p_{tt} I(x), \tag{2}$$

where  $p$  is the polarization in the medium. Let the dielectric susceptibilities of surrounding mediums be the same. That eliminates the Fresnel refraction. The boundary conditions are

$$e(t, x = \pm\infty) = 0, \quad e_t(t, x = \pm\infty) = 0, \tag{3}$$

and the initial conditions are  $e = e_0(x - t)$  following the scattering problem. Then we have

$$e(t = 0, x) = e_0(x), \quad e_t(t = 0, x) = -\partial_x e_0(x). \tag{4}$$

We assume that the initial pulse is located at the left far from the film.

Using the general procedure for solving the wave equation (see [26]), we showed in [27] that the solution of this problem is

$$e(x, t) = e_0(x - t) + \frac{1}{2} \int_{-\infty}^{+\infty} \int_0^t g(y, \tau) [\theta(x - y - t + \tau) - \theta(x - y + t - \tau)] d\tau dy, \tag{5}$$

where we use the step function

$$\theta(z) = \int_{-\infty}^z \delta(x) dx,$$

which can also be written as

$$\theta(z) = \begin{cases} 1 & z > 0, \\ 1/2 & z = 0, \\ 0 & z < 0. \end{cases}$$

When the medium is reduced to a single thin film placed at  $x = a$

$$g(x, t) = -\gamma p_{tt} I(x), \quad \text{where} \quad I(x) = \delta(x - a).$$

The field at  $x = a$  is given by

$$\begin{aligned} e(a, t) &= e_0(a - t) - \frac{\gamma}{2} \int_0^t p_t(a, \tau) [\delta(\tau - t) + \delta(\tau + t)] d\tau \\ &= e_0(a - t) - \gamma p_t(a, t). \end{aligned}$$

With this general formalism, one can address the question of what happens for a medium represented by an ensemble of resonant atoms. These could be molecules, quantum dots, or two- or three-level atoms. Here we will restrict ourselves to the two-level atoms with permanent dipole embedded into a thin film.

## 2.2. The case of a thin film of resonant atoms

Let the plane electromagnetic wave interact with the atoms or molecules characterized by the operator of the dipole transition between resonant energy levels and let this operator have both non-diagonal and diagonal matrix elements [12]. In the two-level approximation the Hamiltonian of the considered model can be written as [15]

$$\hat{H} = \frac{\hbar\omega_0}{2} \begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix} - \begin{pmatrix} d_{11}E & d_{12}E \\ d_{21}E & d_{22}E \end{pmatrix},$$

where  $E$  is the amplitude of the electric field of the electromagnetic wave and  $\omega_0$  is the frequency difference between the levels. The polarization of the medium is

$$P(x, y, t) = l\delta(x - a)n_A p(t), \tag{6}$$

where  $l$  is the film thickness and where  $n_A$  is the volume density of the atoms. We assumed a homogeneous film and neglected the transverse  $y$  dependence of  $P$ . The atomic polarizability  $p$  is given by the expression

$$\begin{aligned} p &= \text{tr}\hat{\rho}\hat{d} = \rho_{11}d_{11} + \rho_{22}d_{22} + \rho_{12}d_{21} + \rho_{21}d_{12} \\ &= \frac{1}{2}(d_{11} + d_{22}) + \frac{1}{2}(d_{11} - d_{22})(\rho_{11} - \rho_{22}) + \rho_{12}d_{21} + \rho_{21}d_{12}, \end{aligned} \tag{7}$$

where  $\hat{\rho}$  is the density matrix and where we used the constraint  $\rho_{11} + \rho_{22} = 1$ . In the expression above, the first term corresponds to the constant polarizability of the molecules. The average of this quantity over all atoms must be zero because we assume no polarization of the medium in the absence of an electromagnetic field. Note that all relaxation processes are neglected because we assume short electromagnetic pulses. We also neglect the dipole–dipole interaction, i.e we assume that the atoms carrying dipoles are rare in the film. The evolution of the elements of  $\hat{\rho}$  is given by the Heisenberg equation  $i\hbar\partial\hat{\rho}/\partial t = \hat{H}\hat{\rho} - \hat{\rho}\hat{H}$  and yields the Bloch equations.

So the total system of equations describing the interaction of an electromagnetic wave with a collection of identical atoms in a film placed at  $x = a$  is [12]

$$\frac{\partial^2 E}{\partial x^2} - \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} = \frac{4\pi n_A}{c^2} l\delta(x - a) \frac{\partial^2}{\partial t^2} \left( \frac{1}{2} (d_{22} - d_{11}) r_3 + d_{12} r_1 \right), \tag{8}$$

$$\frac{\partial r_1}{\partial t} = -[\omega_0 + (d_{11} - d_{22}) E/\hbar] r_2, \tag{9}$$

$$\frac{\partial r_2}{\partial t} = [\omega_0 + (d_{11} - d_{22}) E/\hbar] r_1 + 2(d_{12} E/\hbar) r_3, \tag{10}$$

$$\frac{\partial r_3}{\partial t} = -2(d_{12} E/\hbar) r_2, \tag{11}$$

where  $r_{1,2,3}$  are the components of the Bloch vector:

$$r_1 = \rho_{12} + \rho_{21}, \quad r_2 = -i(\rho_{12} - \rho_{21}), \quad r_3 = \rho_{22} - \rho_{11}. \tag{12}$$

In equations (9)–(11) the field  $E$  is taken in the film, i.e. at  $x = a$ . This system differs from the well-known Maxwell–Bloch equations [28, 29, 30] by the terms containing the parameter  $(d_{11} - d_{22})$ .

We normalize time, space and electric field as

$$\tau = \omega_0 t, \quad \zeta = \frac{\omega_0 x}{c}, \quad e = \frac{2d_{12} E}{\hbar \omega_0}.$$

We also introduce the parameter  $\mu$  which measures the strength of the permanent dipole:

$$\mu = \frac{d_{11} - d_{22}}{2d_{12}}. \tag{13}$$

Then the Maxwell–Bloch equations (8)–(11) take the form

$$r_{1,\tau} = -(1 + \mu e)r_2, \quad r_{2,\tau} = (1 + \mu e)r_1 + er_3, \quad r_{3,\tau} = -er_2, \tag{14}$$

$$e_{,\zeta\zeta} - e_{,\tau\tau} = \alpha(r_1 - \mu r_3)_{,\tau\tau} \delta(\zeta - \zeta_0), \tag{15}$$

where  $\zeta_0 = \omega_0 a/c$ , and

$$\alpha = 8\pi n_A l \frac{d_{12}^2}{\hbar c}. \tag{16}$$

The system can be further simplified by noting from equation (14) that  $(r_1 - \mu r_3)_\tau = -r_2$ . We then get

$$e_{,\tau\tau} - e_{,\zeta\zeta} = \alpha r_{2,\tau} \delta(\zeta - \zeta_0), \tag{17}$$

$$r_{1,\tau} = -(1 + \mu e)r_2, \quad r_{2,\tau} = (1 + \mu e)r_1 + er_3, \quad r_{3,\tau} = -er_2. \tag{18}$$

This is the model that we will analyze in detail in this paper.

We will assume the general initial conditions where the medium is initially at rest so that  $e = 0$ ,  $e_{,\zeta} = e_{,\tau} = 0$ ,  $r_1 = r_2 = 0$ ,  $r_3 = -1$ , at  $\tau \rightarrow -\infty$ . From the Bloch equations (14) we obtain  $(r_1^2 + r_2^2 + r_3^2)_{,\tau} = 0$ , and using the initial conditions we get the value of this integral of motion:

$$r_1^2 + r_2^2 + r_3^2 = 1. \tag{19}$$

We consider the effect of an electromagnetic pulse impinging on the thin film placed at  $\zeta_0 = 0$ . For that we can use the general result (5) to solve the wave equation (17). Taking the right part of (17) as a function  $g(y, \tau)$  under the integral in (5) we obtain the following expression:

$$e(0, \tau) = e_0(-\tau) + \alpha \int_0^\tau r_2(\tau') \delta(\tau - \tau') d\tau' = e_0(-\tau) + \frac{\alpha}{2} r_2(\tau), \tag{20}$$

which represents the strength of the electrical field in the film. Now, using this expression we can write the modified Bloch equations as

$$r_{1,\tau} = -(1 + \mu e_0)r_2 - (\alpha\mu/2)r_2^2, \tag{21}$$

$$r_{2,\tau} = (1 + \mu e_0)r_1 + e_0r_3 + (\alpha\mu/2)r_1r_2 + (\alpha/2)r_2r_3, \tag{22}$$

$$r_{3,\tau} = -e_0r_2 - (\alpha/2)r_2^2. \tag{23}$$

These are the correct equations describing the resonant responses of two-level atoms of a thin film to an ultra-short electromagnetic pulse. Note that the electromagnetic wave is incident normally on the film. Using the equivalent of (20) for  $x \neq 0$  we can write the reflected and transmitted waves in terms of the film variables  $(r_1, r_2, r_3)$ . Second, all atoms of the film are identical. Finally, note that we did not assume any limitation on the time duration of the electromagnetic pulse. It may be a half-period pulse, i.e. an electromagnetic spike or a quasiharmonic wave.

Finally, note that since the motion occurs on the Bloch sphere, the system (23) has the constraint  $r_1^2 + r_2^2 + r_3^2 = 1$ . It is then natural to write it in the reduced coordinates  $(m, \phi)$  such that

$$r_1 = \sqrt{1 - m^2} \cos \phi, \quad r_2 = \sqrt{1 - m^2} \sin \phi, \quad r_3 = m. \tag{24}$$

The system is then

$$m_\tau = -e_0\sqrt{1 - m^2} \sin \phi - \frac{\alpha}{2}(1 - m^2) \sin^2 \phi, \tag{25}$$

$$\phi_\tau = \left( \mu + \frac{m}{\sqrt{1 - m^2}} \cos \phi \right) \left( e_0 + \frac{\alpha}{2}\sqrt{1 - m^2} \sin \phi \right) + 1. \tag{26}$$

### 3. Equilibrium states

We now study the stationary points of the system (23). Initially, before the wave reaches it, the film is at rest. The electromagnetic field  $e_0$  shifts the film state to a new equilibrium. The system then relaxes back to its original state after the wave has passed. We will examine these new transient states and their stability.

For  $e_0$  constant, the system of equations (21)–(23) has the fixed point  $(r_1^*, 0, r_3^*)$  where  $r_{1,3}^*$  satisfy

$$(1 + \mu e_0)r_1^* + e_0r_3^* = 0. \tag{27}$$

Assuming  $r_2^* \neq 0$  leads to a contradiction. From (19) it follows that

$$r_1^{*2} + r_3^{*2} = 1. \tag{28}$$

Combining these two equations we obtain

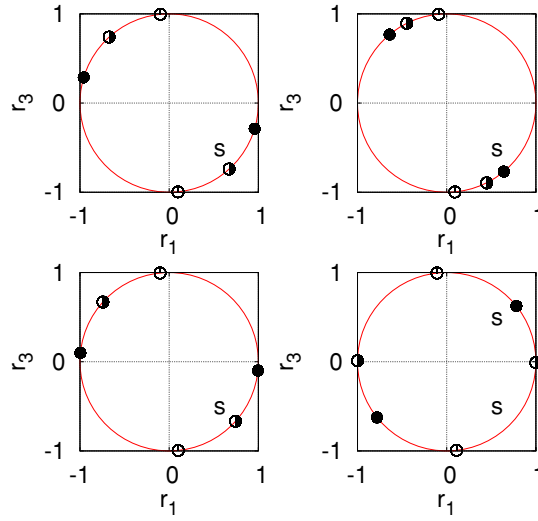
$$r_3^* = \pm \left[ 1 + \frac{e_0^2}{(1 + \mu e_0)^2} \right]^{-1/2}, \quad r_1^* = -\frac{e_0}{1 + \mu e_0}r_3^*. \tag{29}$$

This fixed point corresponds to a stationary polarization and population induced in the two-level atoms by the incident constant field  $e_0$ . When  $e_0 \rightarrow \infty$

$$r_3^* \rightarrow \pm \frac{\mu}{\sqrt{1 + \mu^2}}, \quad r_1^* \rightarrow -\frac{r_3^*}{\mu}. \tag{30}$$

To understand geometrically the position of the fixed points, we can parameterize the Bloch sphere with  $r_2^* = 0$  by

$$r_1^* = \sin \theta, \quad r_3^* = \cos \theta,$$



**Figure 2.** Position of the fixed points  $(r_1^*, 0, r_3^*)$  on the Bloch sphere for four different values of the dipole parameter  $\mu$ , top from left to right  $\mu = 0.1, 1$  and bottom from left to right  $\mu = -0.1, -1$ . The shades correspond to different amplitudes of the electric field  $e_0$ , empty is for  $e_0 = 0.1$ , half-empty is for  $e_0 = 1$  and full is for  $e_0 = 5$ . The stable fixed point region (see the text) is indicated by the letter s.

yielding the relation

$$\tan \theta = -\frac{e_0}{1 + \mu e_0}. \tag{31}$$

The dependence of the values  $r_{1,3}^*$  on the electric field  $e_0$  is monotonic so there is no bistability. The two fixed points are shown in figure 2 for  $\mu = \pm 0.1, \pm 1$ . For  $\mu$  small (left panel) the fixed points start from  $(0, \pm 1)$  and rotate following  $(\sin \theta, \cos \theta)$  with  $\theta$  given by (31). For  $\mu = -1$  and  $e_0 = 1$   $\theta = \pi/2 + n\pi$  so we obtain  $(\pm 1, 0)$ . Since  $r_3$  is the difference in the population of the levels, these are equally populated. When  $\mu$  is large and positive (top right panel) the fixed points do not change very much when the electric field is increased. In contrast, when  $\mu$  is large and negative (bottom right panel), increasing the electric field shifts the fixed points from  $(0, \pm 1)$  to  $(\pm 1, 0)$  for  $e_0 = 1$  and about  $(\pm 1/\sqrt{2}, \pm 1/\sqrt{2})$  for  $e_0 = 5$ . For this last value of the parameter we have a strong population inversion.

To study the stability of the fixed point we linearize the modified Bloch equations. We set  $r_1 = r_1^* + \delta r_1, r_2 = 0 + \delta r_2, r_3 = r_3^* + \delta r_3$ . The linearized modified Bloch equations read

$$\begin{aligned} \delta r_{1\tau} &= -(1 + \mu e_0)\delta r_2, \\ \delta r_{2\tau} &= (1 + \mu e_0)\delta r_1 + e_0\delta r_3 + (\alpha\mu/2)r_1^*\delta r_2 + (\alpha/2)r_3^*\delta r_2, \\ \delta r_{3\tau} &= -e_0\delta r_2, \end{aligned} \tag{32}$$

where as usual the  $\tau$  subscript indicates the derivative. We introduce the field modified frequency

$$\Omega^2 = (1 + \mu e_0)^2 + e_0^2, \tag{33}$$

and ratio

$$b = \frac{\alpha}{4}(\mu r_1^* + r_3^*) = \frac{\alpha}{4} \frac{r_3^*}{1 + \mu e_0} = \pm \frac{\alpha}{4\Omega}. \tag{34}$$



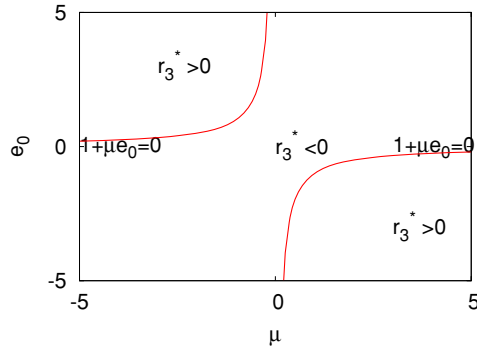


Figure 3. Stability diagram in the  $(\mu, e_0)$  plane indicating which fixed point is stable.

From (32) we can get the equation for  $\delta r_2$ :

$$\delta r_{2\tau\tau} - 2b\delta r_{2,\tau} + \Omega^2\delta r_2 = 0. \tag{35}$$

The characteristic equation is

$$\lambda^2 - 2b\lambda + \Omega^2 = 0,$$

whose roots are

$$\lambda_{1,2} = b \pm i\sqrt{\Omega^2 - b^2} = \pm \frac{\alpha}{4\Omega} \pm i\Omega\sqrt{1 - \frac{\alpha^2}{16\Omega^4}}.$$

Then if  $b < 0$  and  $\Omega^2 > b^2$  the fixed point is stable. Depending on  $\alpha$  two cases occur. Consider first a small  $\alpha$  so that  $\Omega^2 > b^2$ . Then if  $\mu > 0$ ,  $b > 0$  if  $r_3^* > 0$ . Then the fixed point such that  $r_3^* > 0$  is unstable. Conversely the fixed point with  $r_3^* < 0$  is stable. If  $1 + \mu e_0 < 0$  which occurs for  $\mu < 0$  and large fields  $e_0$  the situation is reversed. The fixed point with  $r_3^* > 0$  is stable while the one with  $r_3^* < 0$  is unstable. The oscillation frequency of the orbit as it approaches the fixed point is given by the imaginary part of  $\lambda$ :

$$\omega^* = \Omega\sqrt{1 - \frac{\alpha^2}{16\Omega^4}}. \tag{36}$$

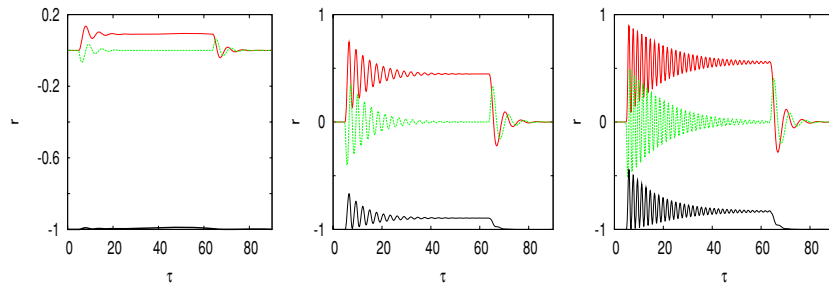
When  $\Omega^2 > b^2$  corresponding to a large  $\alpha$ , there is no imaginary part of  $\lambda$ . Even for very large  $\alpha$  the stability remains unchanged because for the first term  $b = \frac{\alpha}{4\Omega}$  will always dominate the second one  $\Omega\sqrt{\frac{\alpha^2}{16\Omega^4} - 1}$ . Therefore, the fixed point such that  $r_3^* < 0$  is stable (resp. unstable) for  $\mu > 0$  (resp.  $1 + \mu e_0 < 0$ ). In this case there are no oscillations around the fixed point.

To conclude, for all values of  $\alpha$ , the stability is shown in figure 3 in the parameter space  $(\mu, e_0)$ .

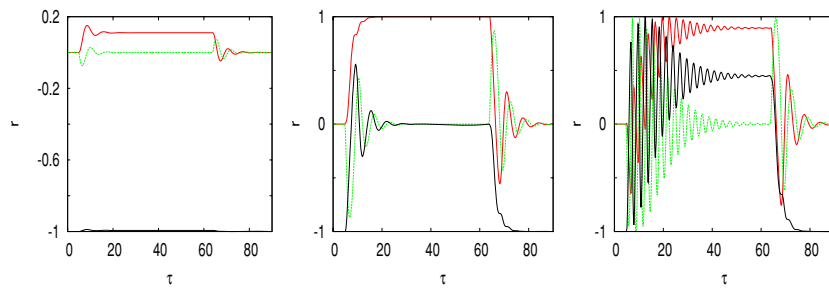
### 3.1. Numerical results

To illustrate the previous analysis, we have solved numerically the system of ordinary differential equations (21)–(23). As a solver we used the Runge–Kutta method of order 4 and 5 enabling step-size control, implemented in the software Dopri5 [31]. For all the runs presented, we choose an incoming pulse  $e_0(x - t)$  where

$$e_0(t) = e_{00} \frac{1}{2} \left[ \tanh\left(\frac{t - t_1}{w}\right) - \tanh\left(\frac{t - t_2}{w}\right) \right], \tag{37}$$



**Figure 4.** Time evolution of the components of the Bloch vector  $(r_1, r_2, r_3)$  in continuous line, long dash and short dash, respectively (red, green and black online). From the left panel to the right panel the amplitude of the field is increased from  $e_0 = 0.1$  (left),  $e_0 = 1$  (middle) to  $e_0 = 2$  (right). The dipole parameter is  $\mu = 1$  and the coupling coefficient is  $\alpha = 1$ .



**Figure 5.** Same as for figure 4 except that the dipole parameter is  $\mu = -1$ .

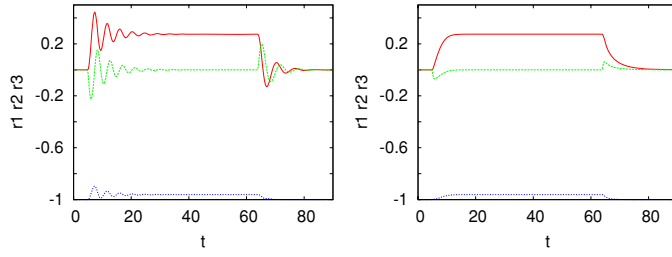
where  $t_1 = -64, t_2 = -5$  and  $w = 0.2$ . In the following we will abusively name the amplitude  $e_{00} e_0$ . We first consider  $\mu = 1$ . figure 4 shows the evolution of the Bloch vector components as a function of time, with from left to right  $e_0 = 0.1, 1$  and  $2$ . As expected the system reaches the stable equilibrium state given by (29) with  $r_3^* < 0$ . The oscillations are given by the frequency  $\omega^* \approx 1.1, 2.2$  and  $3.6$  from left to right. These plots correspond to the upper right panel of figure 2.

The case  $\mu = -1$  is shown in figure 5. When  $e_0$  is small, we have an equilibrium very close to the one for  $\mu = 1$  as expected from the bottom right panel of figure 2. When  $e_0 = 1$  we obtain the situation where  $r_3^* = 0$  and  $r_1^* = \pm 1$ . Note the typical frequency  $\omega^* = 1$  as in the left panel. For a larger field  $e_0 = 2$ , shown in the right panel we obtain as expected an equilibrium  $r_3^* > 0$ . Note the relatively small oscillation frequency  $\omega^* \approx 1.7$  compared to the one for the same  $e_0$  and  $\mu = 1$  (right panel of figure 4).

When  $\alpha$  is large, the fixed point does not change but the eigenvalue of the Jacobian is now real. We then get no oscillations as the system reaches the equilibrium as shown in the right panel of figure 6 for which  $e_0 = 0.4, \mu = 1$  and  $\alpha = 10$ . Reducing  $\alpha$  to 1 restores the oscillations of frequency  $\omega^*$  as shown in the left panel of figure 6.

#### 4. Spectroscopic analysis of the film

We now assume that the film is irradiated by a long pulse  $e_0$  on top of which we add a small harmonic wave  $\delta e$ . The duration of  $e_0$  is chosen large compared to  $1/\omega_0$  but smaller than the



**Figure 6.** Time evolution of the components of the Bloch vector  $(r_1, r_2, r_3)$  in continuous line, long dash and short dash, respectively (red, green and black online) for two values of the coupling parameter  $\alpha = 1$  for the left plot and  $\alpha = 10$  for the right plot. The amplitude of the electric field is  $e_0 = 0.4$ . All the other parameters are as in figure 4.

relaxation time of the medium. The small wave is applied after any transient due to  $e_0$  has disappeared. Then the film has settled in its stable fixed point and its behavior can be described by its linearization. It is then possible to examine how small waves of different frequencies get scattered by the film. Using this spectroscopic analysis of the film, we show that one can recover the atomic dipolar parameter  $\mu$  and the coupling parameter  $\alpha$ .

For a general thin layer medium, the scattering theory cannot be solved except in particular cases. One example is the Schroedinger equation with point nonlinearities studied by Malomed and Azbel [32]. Because of the cubic nonlinearity, they were able to obtain the scattering solution and study its modulational stability. Also Kaup and Malomed [33] studied the thin layer three-wave mixing system. Here the scattering theory was solved using approximations. These difficulties are absent in our case because our system is linear and the scattering theory can be set up clearly.

To do this, first we linearize the system (17), (18) assuming that  $e_0$  is constant. We get

$$\delta e_{\tau\tau} - \delta e_{\zeta\zeta} = \alpha \delta_D(\zeta) \delta r_{2,\tau}, \tag{38}$$

$$\delta r_{1,\tau} = -(1 + \mu e_0) \delta r_2, \tag{39}$$

$$\delta r_{2,\tau} = (1 + \mu e_0) \delta r_1 + e_0 \delta r_3 + (\mu r_1^* + r_3^*) \delta e, \tag{40}$$

$$\delta r_{3,\tau} = -e_0 \delta r_2, \tag{41}$$

where we temporarily use  $\delta_D$  to define the Dirac delta function. We separate time and space,

$$\delta e = E(\zeta) e^{-i\omega\tau}, \quad \delta r_{1,2,3} = R_{1,2,3} e^{-i\omega\tau}, \tag{42}$$

and obtain the system

$$E_{\zeta\zeta} + \omega^2 E = i\omega\alpha \delta_D(\zeta) R_2, \tag{43}$$

$$i\omega R_1 = (1 + \mu e_0) R_2, \tag{44}$$

$$-i\omega R_2 = (1 + \mu e_0) R_1 + e_0 R_3 + (\mu r_1^* + r_3^*) E, \tag{45}$$

$$i\omega R_3 = e_0 R_2. \tag{46}$$

From the second and fourth equations we get

$$R_1 = -i \frac{(1 + \mu e_0)}{\omega} R_2, \quad R_3 = -i \frac{e_0}{\omega} R_2. \tag{47}$$

Plugging these expression into (43) results in

$$R_2 = i \frac{(\mu r_1^* + r_3^*)\omega E}{\omega^2 - \Omega^2}, \tag{48}$$

where  $\Omega$  is given by (33). The substitution of the previous equation into (38) leads to the non-standard eigenvalue problem

$$E_{xx} + \omega^2 \left[ 1 + \frac{\alpha(\mu r_1^* + r_3^*)\delta_D(x)}{\omega^2 - \Omega^2} \right] E = 0. \tag{49}$$

As usual we assume a scattering experiment so that the field is given by

$$E = \begin{cases} A e^{i\omega x} + B e^{-i\omega x}, & x < 0, \\ C e^{i\omega x}, & x > 0. \end{cases} \tag{50}$$

At the film  $x = 0$  the field is continuous and its gradient satisfies the jump condition

$$\begin{cases} E(0+) = E(0-) = E(0) \\ E_x(0+) - E_x(0-) + \beta E(0) = 0, \end{cases} \tag{51}$$

where

$$\beta = \frac{\alpha(\mu r_1^* + r_3^*)\omega^2}{\omega^2 - \Omega^2}.$$

Writing the two conditions (51) using the left and right fields results in

$$\begin{cases} A + B = C \\ i\omega(C - A + B) + \beta C = 0. \end{cases} \tag{52}$$

The solution is

$$B = \frac{i\beta}{2\omega} \frac{1}{1 + \frac{i\beta}{2\omega}} A, \quad C = \frac{1}{1 + \frac{i\beta}{2\omega}} A.$$

The fixed point is such that  $(1 + \mu e_0)r_1^* + e_0 r_3^* = 0$ , so that  $\mu r_1^* + r_3^* = -\frac{r_1^*}{e_0}$ , so the reflection and transmission coefficients are

$$R = \frac{B}{A} = -\frac{i\alpha\omega(r_1^*/e_0)}{2(\omega^2 - \Omega^2) + i\alpha\omega(r_1^*/e_0)} \tag{53}$$

$$T = \frac{C}{A} = \frac{(\omega^2 - \Omega^2)}{(\omega^2 - \Omega^2) + i\alpha\omega(r_1^*/2e_0)} \tag{54}$$

where the reflection and transmission coefficients satisfy  $|R|^2 + |T|^2 = 1$  From (29) we have

$$r_1^*/e_0 = \pm 1/\Omega, \tag{55}$$

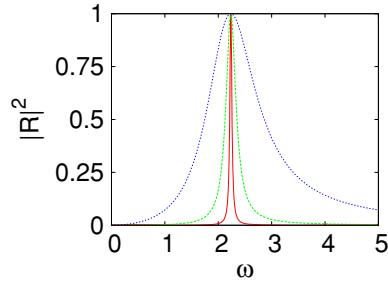
so that the transmission coefficient is

$$T = \frac{2\Omega(\omega^2 - \Omega^2)}{2\Omega(\omega^2 - \Omega^2) \pm i\alpha\omega}. \tag{56}$$

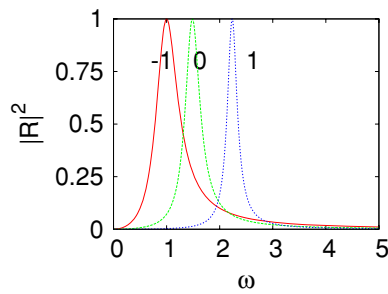
The modulus squared of the transmission and reflection coefficients are then

$$|T|^2 = \frac{4\Omega^2(\omega^2 - \Omega^2)^2}{4\Omega^2(\omega^2 - \Omega^2)^2 + \alpha^2\omega^2}, \tag{57}$$

$$|R|^2 = \frac{\alpha^2\omega^2}{4\Omega^2(\omega^2 - \Omega^2)^2 + \alpha^2\omega^2}. \tag{58}$$



**Figure 7.** Square of the modulus  $|R|^2$  of the reflection coefficient as a function of the frequency  $\omega$  for three different values of the coupling coefficient  $\alpha = 0.2$  (continuous line, red online), 1 (long dashed line, green online) and 5 (short dashed line, blue online). The parameters are  $\mu = 1, e_0 = 1$ .



**Figure 8.** Square of the modulus  $|R|^2$  of the reflection coefficient as a function of the frequency  $\omega$  for three different values of the dipole parameter  $\mu = -1, 0.1$  and 1 as indicated in the plot. The parameters are  $e_0 = 1, \alpha = 1$ .

Let us examine how  $|R|^2$  depends on the different parameters. It attains its maximum 1 for  $\omega = \Omega$  and decays at infinity as  $\frac{1}{\omega^2}$ . The half-width of the resonance, such that  $|R|^2(\omega_h) = 1/2$  can be easily obtained. Solving the quadratic equation for  $\omega_h^2$  we get

$$\omega_h^2 = \Omega^2 \pm \frac{\alpha}{2} \sqrt{1 + \frac{\alpha^2}{16\Omega^4}} + \frac{\alpha^2}{8\Omega^2} \approx \Omega^2 \pm \frac{\alpha}{2}, \tag{59}$$

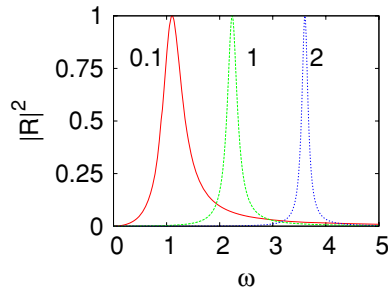
for large  $\Omega$ . Therefore, the half-width is

$$\omega_h \approx \Omega \pm \frac{\alpha}{4\Omega}. \tag{60}$$

Figure 7 shows the square of the modulus  $|R|^2$  for a fixed  $\Omega$  ( $e_0 = 1, \mu = 1$ ). As expected the half-width is proportional to  $\alpha$ . Note how the resonance becomes asymmetric for large  $\alpha$  indicating that all higher order terms in (59) should be considered.

When  $\mu$  is varied  $\Omega$  varies so  $|R|^2$  will be shifted. Figure 8 shows  $|R|^2$  for  $\mu = -1, 0$  and 1. As expected for  $\mu < 0$  (resp.  $\mu > 0$ ) the resonance is shifted toward low (resp. high) frequencies. For  $\mu = -1$ , the higher order terms in (59) should be taken into account and the resonance is not symmetric. When  $\mu = 1$ , the higher order terms can be dropped and the resonance curve becomes symmetric.

The amplitude of the incoming pulse  $e_0$  will also change the resonance by shifting  $\Omega$ . Figure 9 shows  $|R|^2$  for  $e_0 = 0.1, 1$  and 2. As expected, for  $e_0 = 0.1$  the resonance is asymmetric. It becomes symmetric for  $e_0 \geq 1$ .



**Figure 9.** Square of the modulus  $|R|^2$  of the reflection coefficient as a function of the frequency  $\omega$  for three different values of the field amplitude  $e_0 = 0.1, 1$  and  $2$ . The parameters are  $\mu = 1, \alpha = 1$ .

The reflection curve  $R(\omega)$  allows us to estimate the atomic parameter  $\mu$  and coupling parameter  $\alpha$  through formulas (33) and (59). The position of the resonance gives  $\Omega$  and from there one can compute  $\mu$ . We have

$$\mu = -\frac{1}{e_0} \pm \frac{\sqrt{\Omega^2 - e_0^2}}{e_0}. \tag{61}$$

The coupling parameter  $\alpha$  can then be obtained from (59).

### 5. Conclusion

We consider the propagation of an electromagnetic pulse through a thin film containing two-level atoms with permanent dipole. Therefore, there are both non-diagonal and diagonal matrix elements of the operator of the dipole transition between resonant energy levels. The exact solution of the linear wave equation allows us to derive the modified Bloch equations. Thus, the problem of propagating extremely short (one- or few-cycle) pulses through a thin film is reduced to the analysis of a system of nonlinear ordinary differential equations on the Bloch sphere.

In the presence of a constant background field the equilibrium state of the system film/field is changed. These new equilibrium states are the fixed points of the modified Bloch equations. They depend on the field amplitude, the difference of diagonal elements of the operator of the dipole transition (dipole parameter) and the coupling constant. The stability analysis of these fixed points indicates which states will be attained by the system. For the stable states the population difference is negative or positive depending on the sign of the dipole parameter. When the film is illuminated by an electromagnetic field it reaches the new stable state with a typical relaxation time which depends on the field amplitude, the dipole parameter and the coupling constant. Using our estimate, experimentalists could measure the dipole parameter.

In the last part of the paper we considered that in addition to a constant background, the thin film is irradiated by a small harmonic field of frequency  $\omega$ . This spectroscopy analysis yields the reflection and transmission coefficients of the film as a function of  $\omega$ . As expected the film is completely opaque, i.e. its reflection coefficient is equal to 1 for the field modified transition frequency  $\Omega$  given by (33). We found that the width of the resonant curve is proportional to the coupling constant. The position of the resonance depends on the dipole parameter and the ground field.

The modern progress in nano-technology allows us to produce thin films of different features and the investigation of such features is attractive. Our study shows that the fast

electromagnetic response of a thin film could be used in experiments to measure intrinsic parameters of generalized atoms (quantum dots, meta-atoms, molecules . . .) and their coupling parameter to the field.

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