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*Lecture notes*

# LECTURE 12

In this final lecture, we will study the behaviour of the Bloch equations in different regimes of resonance and relaxation. The Bloch equations are formulated as a vector model, and numerical solutions to the equations are discussed.

For steady-state interaction, the polarization density of the medium, as obtained from the Bloch equations, is expressed in a closed form. The closed solution is then expanded in a power series, which when compared with the series obtained from the susceptibility formalism finally tie together the Bloch theory with the susceptibilities.

The outline for this lecture is:

- Recapitulation of the Bloch equations
- The vector model of the Bloch equations
- Special cases and examples
- Steady-state regime
- The intensity dependent refractive index at steady-state
- Comparison with the susceptibility model

## Recapitulation of the Bloch equations for two-level systems

Assuming two states $|a\rangle$ and $|b\rangle$ to be sufficiently similar in order for their respective lifetimes $T_a \approx T_b \approx T_1$ to hold, where $T_1$ is the *longitudinal relaxation time*, the Bloch equations for the two-level are given as

$$\frac{du}{dt} = -\Delta v - u/T_2, \tag{1a}$$

$$\frac{dv}{dt} = \Delta u + \beta(t)w - v/T_2, \tag{1b}$$

$$\frac{dw}{dt} = -\beta(t)v - (w - w_0)/T_1, \tag{1c}$$

where $\beta \equiv e r^{\alpha}_{ab} E^{\alpha}_{\omega}(t)/\hbar$ is the Rabi frequency, being a quantity linear in the applied electric field of the light, $\Delta \equiv \Omega_{ba} - \omega$ is the detuning of the angular frequency of the light from the transition frequency $\Omega_{ba} \equiv (\mathbb{E}_b - \mathbb{E}_a)/\hbar$, and where the variables $(u, v, w)$ are related to the matrix elements $\rho_{mn}$ of the density operator as

$$u = \rho^{\Omega}_{ba} + \rho^{\Omega}_{ab},$$
$$v = i(\rho^{\Omega}_{ba} - \rho^{\Omega}_{ab}),$$
$$w = \rho_{bb} - \rho_{aa}.$$

In these equations, $\rho^{\Omega}_{ab}$ is the *temporal envelope of the off-diagonal elements*, given by

$$\rho_{ab} \equiv \rho^{\Omega}_{ab} \exp[i(\Omega_{ba} - \Delta)t].$$

In the Bloch equations (1), the variable $w$ describes the population inversion of the two-level system, while $u$ and $v$ are related to the dispersive and absorptive components of the polarization density of the medium. In the Bloch equations, $w_0 \equiv \rho_0(b) - \rho_0(a)$ is the thermal equilibrium inversion of the system with no optical field applied.

**The resulting electric polarization density of the medium**

The so far developed theory of the density matrix under resonant interaction can now be applied to the calculation of the electric polarization density of the medium, consisting of $N$ identical molecules per unit volume, as

$$\begin{aligned}
P_\mu(\mathbf{r},t) &= N\langle e\hat{r}_\mu\rangle \\
&= N\,\mathrm{Tr}[\hat{\rho}e\hat{r}_\mu] \\
&= N\sum_{k=a,b}\langle k|\hat{\rho}e\hat{r}_\mu|k\rangle \\
&= N\sum_{k=a,b}\sum_{j=a,b}\langle k|\hat{\rho}|j\rangle\langle j|e\hat{r}_\mu|k\rangle \\
&= N\sum_{k=a,b}\{\langle k|\hat{\rho}|a\rangle\langle a|e\hat{r}_\mu|k\rangle + \langle k|\hat{\rho}|b\rangle\langle b|e\hat{r}_\mu|k\rangle\} \\
&= N\{\langle a|\hat{\rho}|a\rangle\langle a|e\hat{r}_\mu|a\rangle + \langle b|\hat{\rho}|a\rangle\langle a|e\hat{r}_\mu|b\rangle + \langle a|\hat{\rho}|b\rangle\langle b|e\hat{r}_\mu|a\rangle + \langle b|\hat{\rho}|b\rangle\langle b|e\hat{r}_\mu|b\rangle\} \\
&= N(\rho_{ba}er^\mu_{ab} + \rho_{ab}er^\mu_{ba}) \\
&= \{\text{Make use of } \rho_{ab} = (u+iv)\exp(i\omega t) = \rho^*_{ba}\} \\
&= N[(u-iv)\exp(-i\omega t)er^\mu_{ab} + (u+iv)\exp(i\omega t)er^\mu_{ba}].
\end{aligned}$$

The temporal envelope $P^\mu_\omega$ of the polarization density is throughout this course as well as in Butcher and Cotter's book taken as

$$P^\mu(\mathbf{r},t) = \mathrm{Re}[P^\mu_\omega \exp(-i\omega t)],$$

and by identifying this expression with the right-hand side of the result above, we hence finally have obtained the polarization density in terms of the Bloch parameters $(u,v,w)$ as

$$P^\mu_\omega(\mathbf{r},t) = Ner^\mu_{ab}(u-iv). \tag{2}$$

This expression for the temporal envelope of the polarization density is exactly in the same mode of description as the one as previously used in the susceptibility theory, as in the wave equations developed in lecture eight. The only difference is that now we instead consider the polarization density as given by a non-perturbative analysis. Taken together with the Maxwell's equations (or the propern wave equation for the envelopes of the fields), the Bloch equations are known as the *Maxwell–Bloch equations*.

From Eq. (2), it should now be clear that the Bloch variable $u$ essentially gives the in-phase part of the polarization density (at least in this case, where we may consider the transition dipole moments to be real-valued), corresponding to the dispersive components of the interaction between light and matter, while the Bloch variable $v$ on the other hand gives terms which are shifted ninety degrees out of phase with the optical field, hence corresponding to absorptive terms.

**The vector model of the Bloch equations**

In the form of Eqs. (1), the Bloch equations can be expressed in the form of an Euler equation as

$$\frac{d\mathbf{R}}{dt} = \mathbf{\Omega} \times \mathbf{R} - \underbrace{(u/T_2, v/T_2, (w - w_0)/T_1)}_{\text{relaxation term}}, \qquad \text{[B. \& C. (6.54)]}$$

where $\mathbf{R} = (u, v, w)$ is the so-called *Bloch vector*, that in the abstract $(\mathbf{e}_u, \mathbf{e}_v, \mathbf{e}_w)$-space describes the state of the medium, and

$$\mathbf{\Omega} = (-\beta(t), 0, \Delta)$$

is the vector that gives the precession of the Bloch vector (see Fig. 1).

This form, originally proposed in 1946 by Felix Bloch[1] for the motion of a nuclear spin in a magnetic field under influence of radio-frequency electromagnetic fields, and later on adopted by Feynman, Vernon, and Hellwarth[2] for solving problems in maser theory[3], corresponds to the motion of a damped gyroscope in the presence of a gravitational field. In this analogy, the vector $\mathbf{\Omega}$ can be considered as the torque vector of the spinning top of the gyroscope.

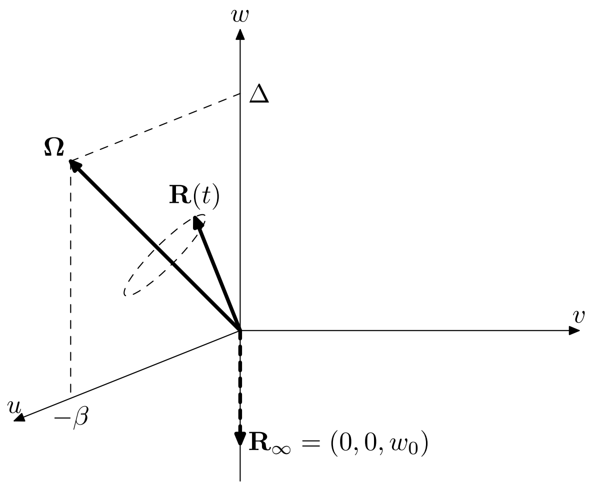


Figure 1. Evolution of the Bloch vector $\mathbf{R}(t) = (u(t), v(t), w(t))$ around the "torque vector" $\mathbf{\Omega} = (-\beta(t), 0, \Delta)$. In the absence of optical fields, the Bloch vector relax towards the thermal equilibrium state $\mathbf{R}_\infty = (0, 0, w_0)$, where $w_0 = \rho(b) - \rho(a)$ is the molecular population inversion at thermal equilibrium. At moderate temperatures, the thermal equilibrium population inversion is very close to $w_0 = -1$.

From the vector form of the Bloch equations, it is found that the Bloch vector rotates around the torque vector $\mathbf{\Omega}$ as the state of matter approaches steady state. For an adiabatically changing applied optical field (i. e. a slowly varying envelope of the field), this precession follows the torque vector.

The relaxation term in the vector Bloch equations also tells us that the relaxation along the $w$-direction is given by the time constant $T_1$, while the relaxation in the $(u, v)$-plane instead is given by the time constant $T_2$. By considering the $w$-axis as the "longitudinal" direction and the $(u, v)$-plane as the "transverse" plane, the terminology for $T_1$ as being the "longitudinal relaxation time" and $T_2$ as being the "transverse relaxation time" should hence be clear.

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[1] F. Bloch, *Nuclear induction*, Phys. Rev. **70**, 460 (1946). Felix Bloch was in 1952 awarded the Nobel prize in physics, together with Edward Mills Purcell, "for their development of new methods for nuclear magnetic precision measurements and discoveries in connection therewith".

[2] R. P. Feynman, F. L. Vernon, and R. W. Hellwarth, *Geometrical representation of the Schrödinger equation for solving maser problems*, J. Appl. Phys. **28**, 49 (1957).

[3] Microwave Amplification by Stimulated Emission of Radiation, a device for amplification of microwaves, essentially working on the same principle as the laser.

**Transient build-up at exact resonance as the optical field is switched on**

*The case $T_1 \gg T_2$ – Longitudinal relaxation slower than transverse relaxation*

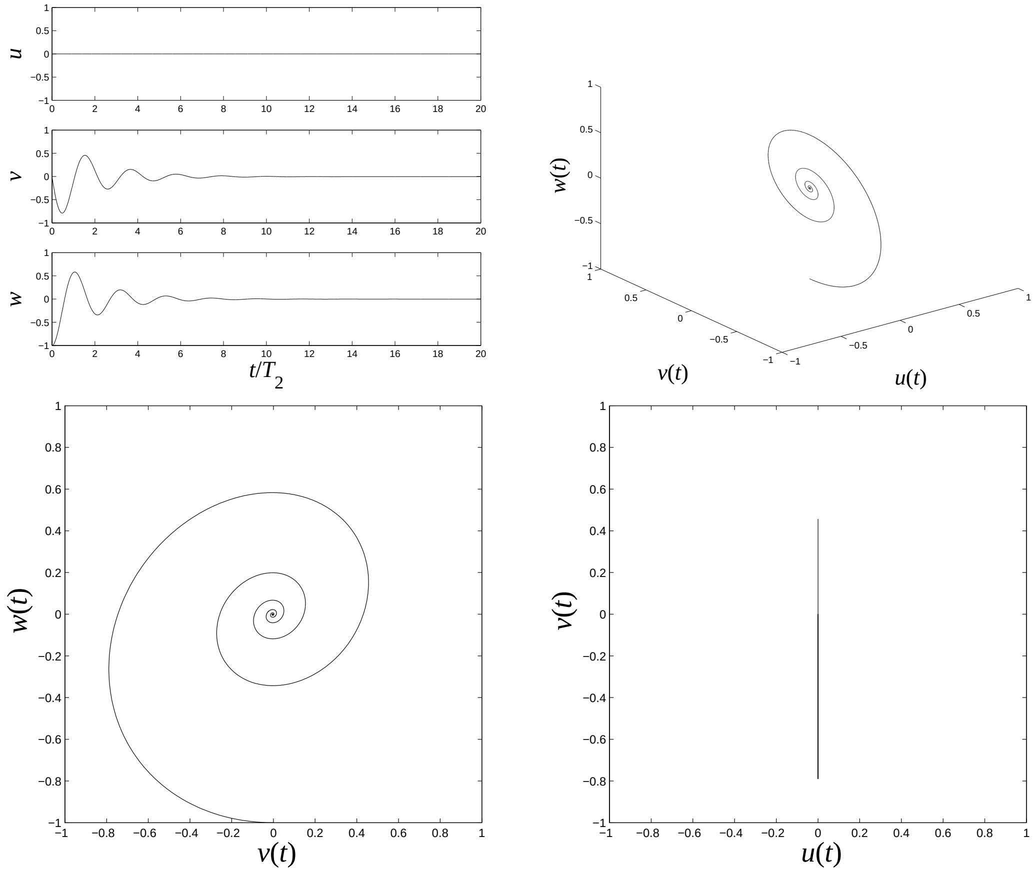


Figure 2a. Evolution of the Bloch vector $(u(t), v(t), w(t))$ as the optical field is switched on, for the exactly resonant case ($\delta = 0$), and with the longitudinal relaxation time being much greater than the transverse relaxation time ($T_1 \gg T_2$). The parameters used in the simulation are $\eta \equiv T_1/T_2 = 100$, $\delta \equiv \Delta T_2 = 0$, $w_0 = -1$, and $\gamma(t) \equiv \beta(t)T_2 = 3$, $t > 0$. The medium was initially at thermal equilibrium, $(u(0), v(0), w(0)) = (0, 0, w_0) = -(0, 0, 1)$.

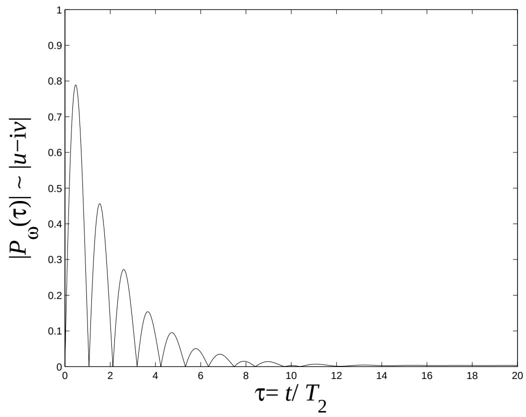


Figure 2b. Evolution of the magnitude of the polarization density $|P_\omega(t)| \sim |u(t) - iv(t)|$ as the optical field is switched on, corresponding to the simulation shown in Fig. 2a.

*The case $T_1 \approx T_2$ – Longitudinal relaxation approximately equal to transverse relaxation*

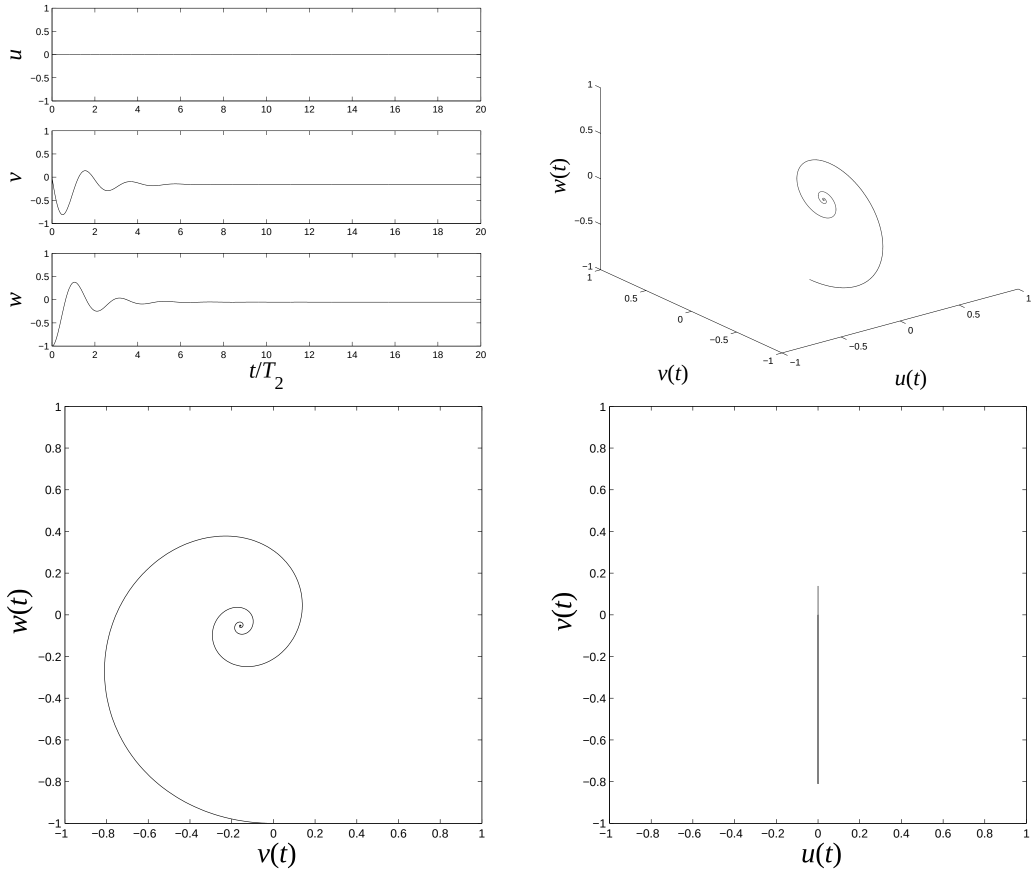


Figure 3a. Evolution of the Bloch vector $(u(t), v(t), w(t))$ as the optical field is switched on, for the exactly resonant case ($\delta = 0$), and with the longitudinal relaxation time being approximately equal to the transverse relaxation time ($T_1 \approx T_2$). The parameters used in the simulation are $\eta \equiv T_1/T_2 = 2$, $\delta \equiv \Delta T_2 = 0$, $w_0 = -1$, and $\gamma(t) \equiv \beta(t)T_2 = 3$, $t > 0$. The medium was initially at thermal equilibrium, $(u(0), v(0), w(0)) = (0, 0, w_0) = -(0, 0, 1)$.

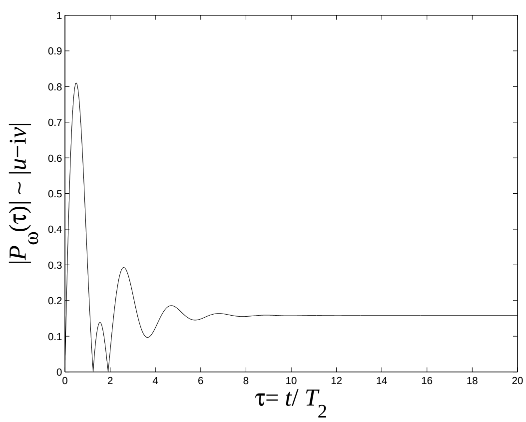


Figure 3b. Evolution of the magnitude of the polarization density $|P_\omega(t)| \sim |u(t) - iv(t)|$ as the optical field is switched on, corresponding to the simulation shown in Fig. 3a.

**Transient build-up at off-resonance as the optical field is switched on**

*The case $T_1 \approx T_2$ – Longitudinal relaxation approximately equal to transverse relaxation*

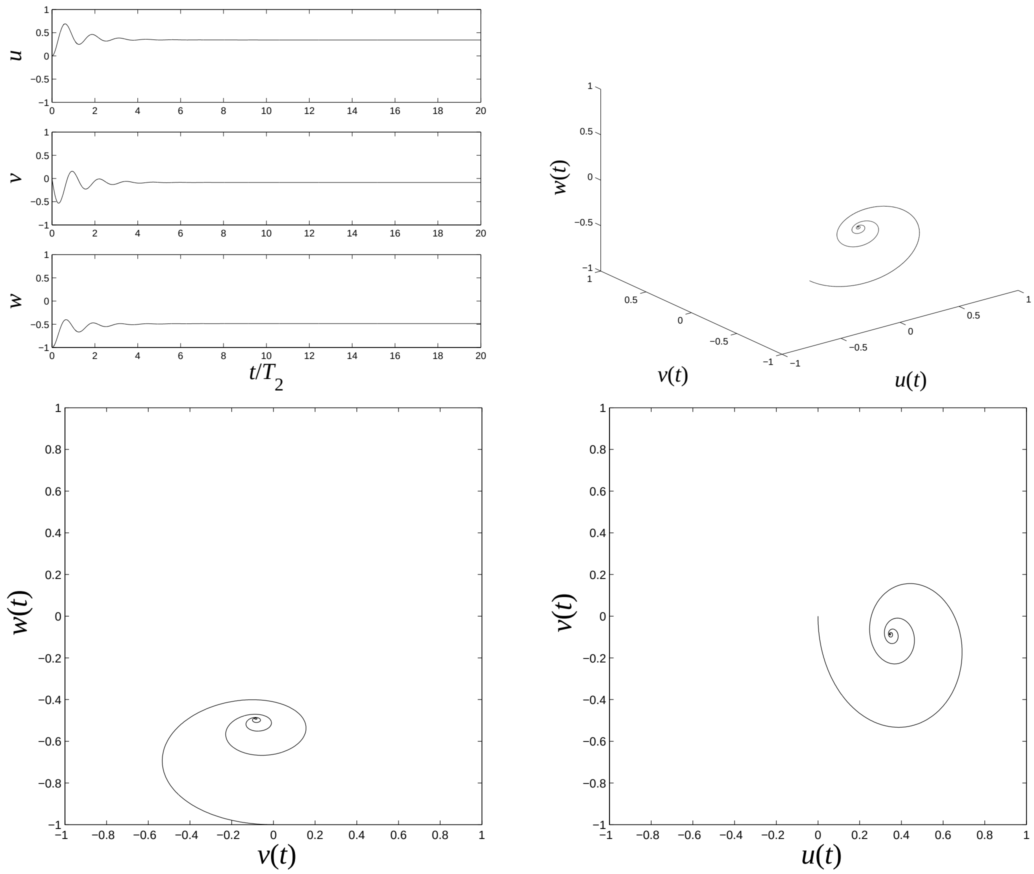


Figure 4a. Evolution of the Bloch vector $(u(t), v(t), w(t))$ as the optical field is switched on, for the off-resonant case ($\delta \neq 0$), and with the longitudinal relaxation time being approximately equal to the transverse relaxation time ($T_1 \approx T_2$). The parameters used in the simulation are $\eta \equiv T_1/T_2 = 2$, $\delta \equiv \Delta T_2 = 4$, $w_0 = -1$, and $\gamma(t) \equiv \beta(t)T_2 = 3$, $t > 0$. The medium was initially at thermal equilibrium, $(u(0), v(0), w(0)) = (0, 0, w_0) = -(0, 0, 1)$.

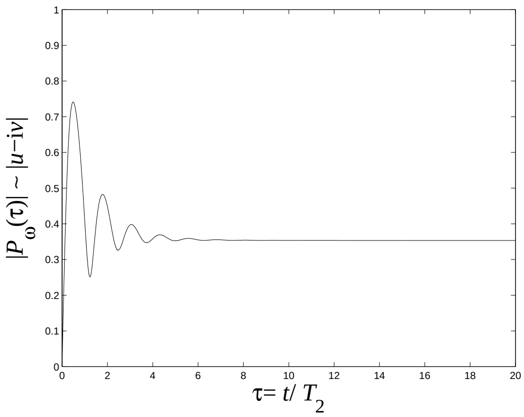


Figure 4b. Evolution of the magnitude of the polarization density $|P_\omega(t)| \sim |u(t) - iv(t)|$ as the optical field is switched on, corresponding to the simulation shown in Fig. 4a.

**Transient decay for a process tuned to exact resonance**

*The case $T_1 \gg T_2$ – Longitudinal relaxation slower than transverse relaxation*

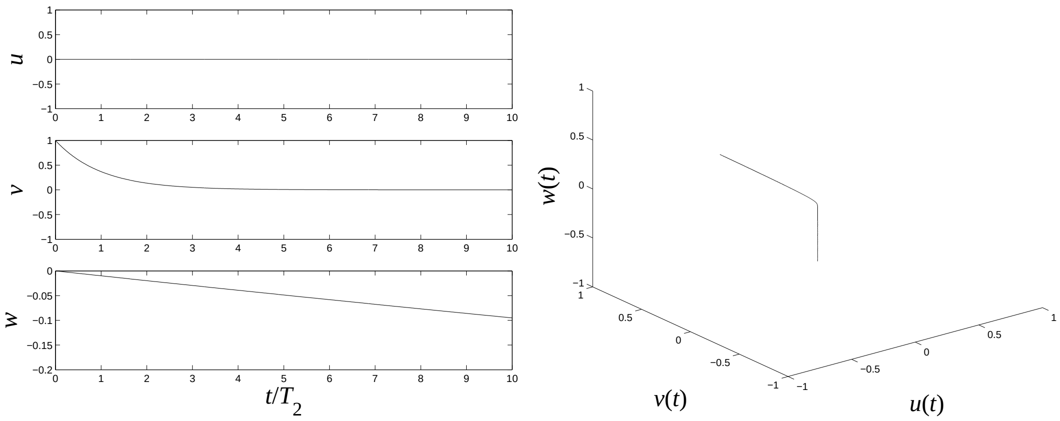


Figure 5. Evolution of the Bloch vector $(u(t), v(t), w(t))$ after the optical field is switched off, for the case of tuning to exact resonance ($\delta = 0$), and with the longitudinal relaxation time being much greater than the transverse relaxation time ($T_1 \gg T_2$). The parameters used in the simulation are $\eta \equiv T_1/T_2 = 100$, $\delta \equiv \Delta T_2 = 0$, $w_0 = -1$, and $\gamma(t) \equiv \beta(t)T_2 = 0$.

*The case $T_1 \approx T_2$ – Longitudinal relaxation approximately equal to transverse relaxation*

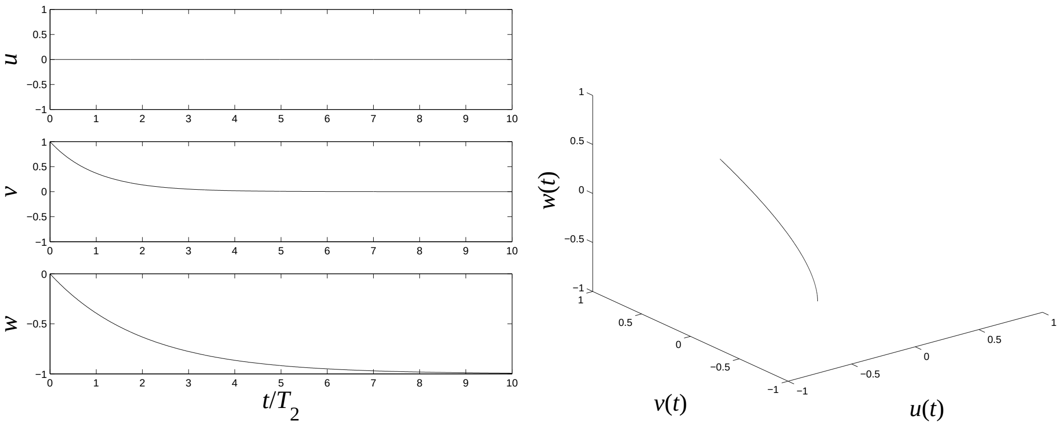


Figure 6. Evolution of the Bloch vector $(u(t), v(t), w(t))$ after the optical field is switched off, for the case of tuning to exact resonance ($\delta = 0$), and with the longitudinal relaxation time being approximately equal to the transverse relaxation time ($T_1 \approx T_2$). The parameters used in the simulation are $\eta \equiv T_1/T_2 = 2$, $\delta \equiv \Delta T_2 = 0$, $w_0 = -1$, and $\gamma(t) \equiv \beta(t)T_2 = 0$.

**Transient decay for a slightly off-resonant process**

*The case $T_1 \gg T_2$ – Longitudinal relaxation slower than transverse relaxation*

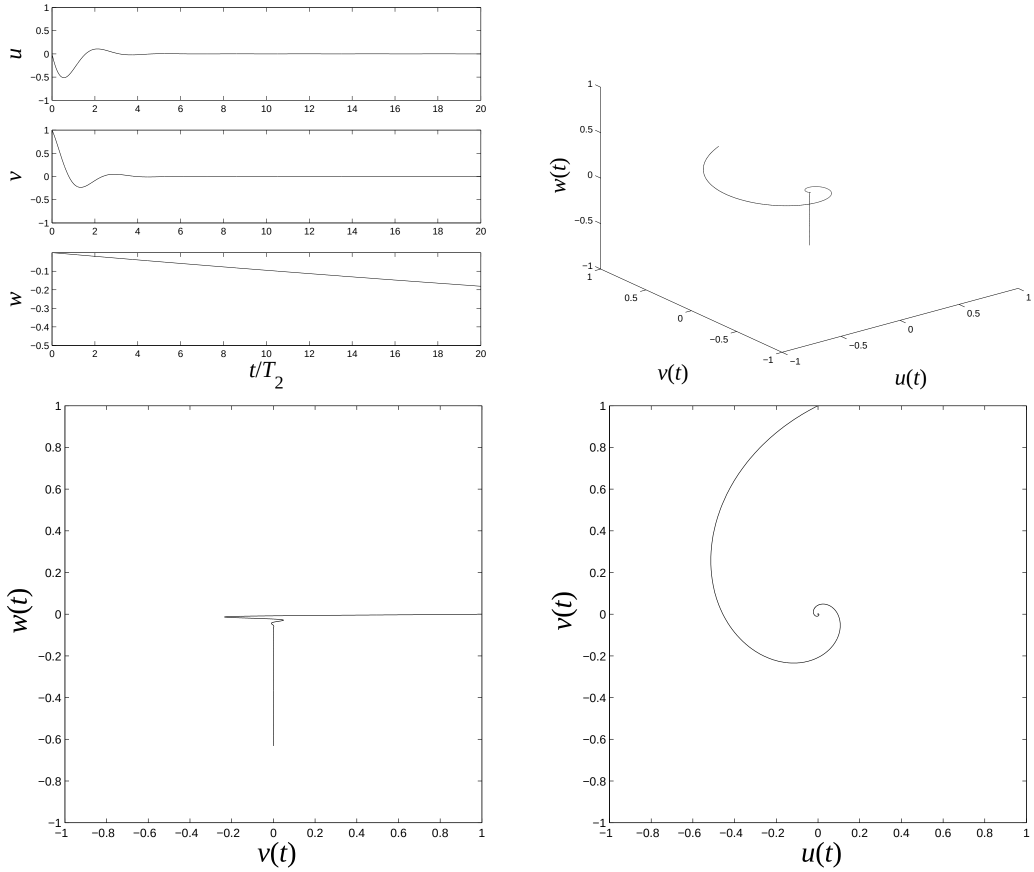


Figure 7a. Evolution of the Bloch vector $(u(t), v(t), w(t))$ after the optical field is switched off, for the off-resonant case ($\delta \neq 0$), and with the longitudinal relaxation time being much greater than the transverse relaxation time ($T_1 \gg T_2$). The parameters used in the simulation are $\eta \equiv T_1/T_2 = 100$, $\delta \equiv \Delta T_2 = 2$, $w_0 = -1$, and $\gamma(t) \equiv \beta(t)T_2 = 0$. (Compare with Fig. 5 for the exactly resonant case.)

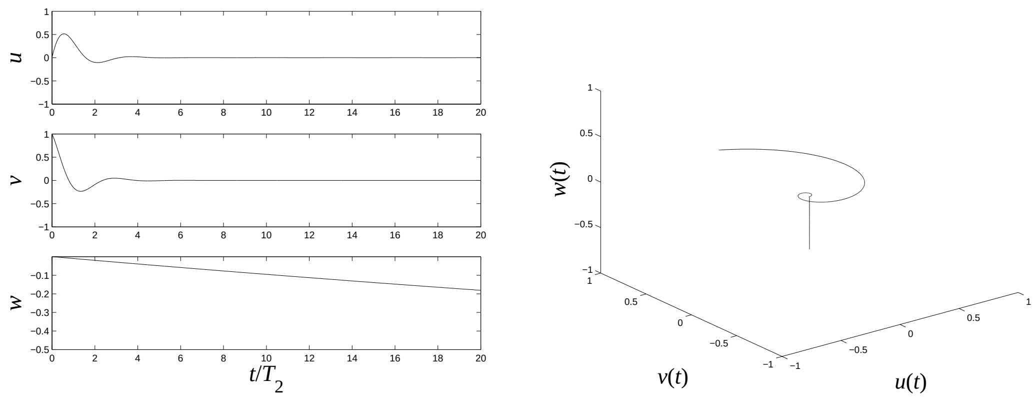


Figure 7b. Same as Fig. 7a, but with $\delta = -2$ as negative.

*The case $T_1 \approx T_2$ – Longitudinal relaxation approximately equal to transverse relaxation*

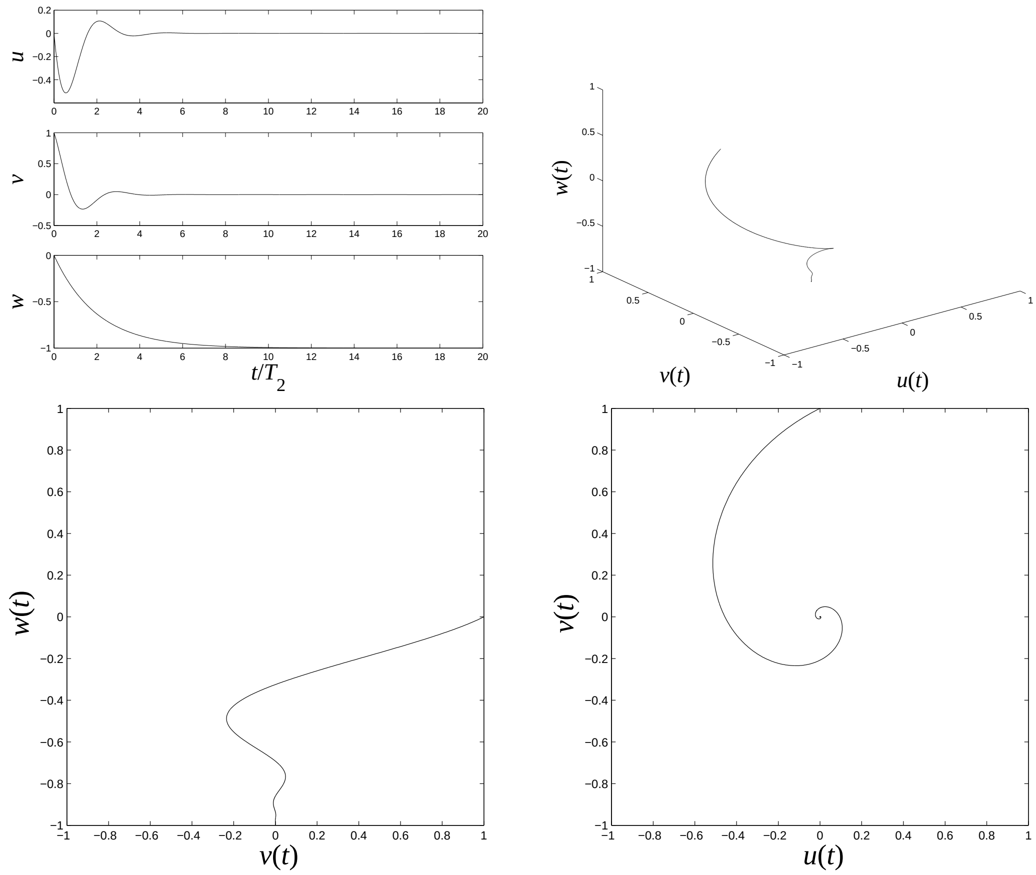


Figure 8a. Evolution of the Bloch vector $(u(t), v(t), w(t))$ after the optical field is switched off, for the off-resonant case ($\delta \neq 0$), and with the longitudinal relaxation time being approximately equal to the transverse relaxation time ($T_1 \approx T_2$). The parameters used in the simulation are $\eta \equiv T_1/T_2 = 2$, $\delta \equiv \Delta T_2 = 2$, $w_0 = -1$, and $\gamma(t) \equiv \beta(t)T_2 = 0$. (Compare with Fig. 6 for the exactly resonant case.)

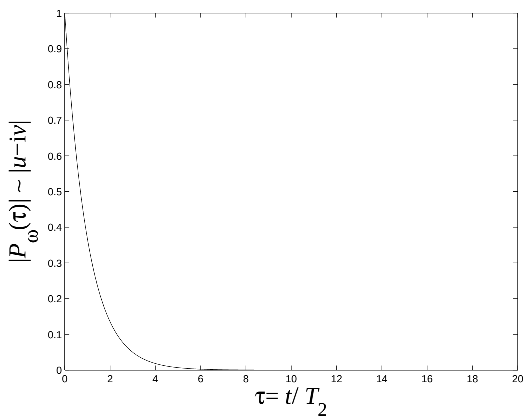


Figure 8b. Evolution of the magnitude of the polarization density $|P_\omega(t)| \sim |u(t) - iv(t)|$ as the optical field is switched on, corresponding to the simulation shown in Fig. 8a.

**Transient decay for a far off-resonant process**

*The case $T_1 \gg T_2$ – Longitudinal relaxation slower than transverse relaxation*

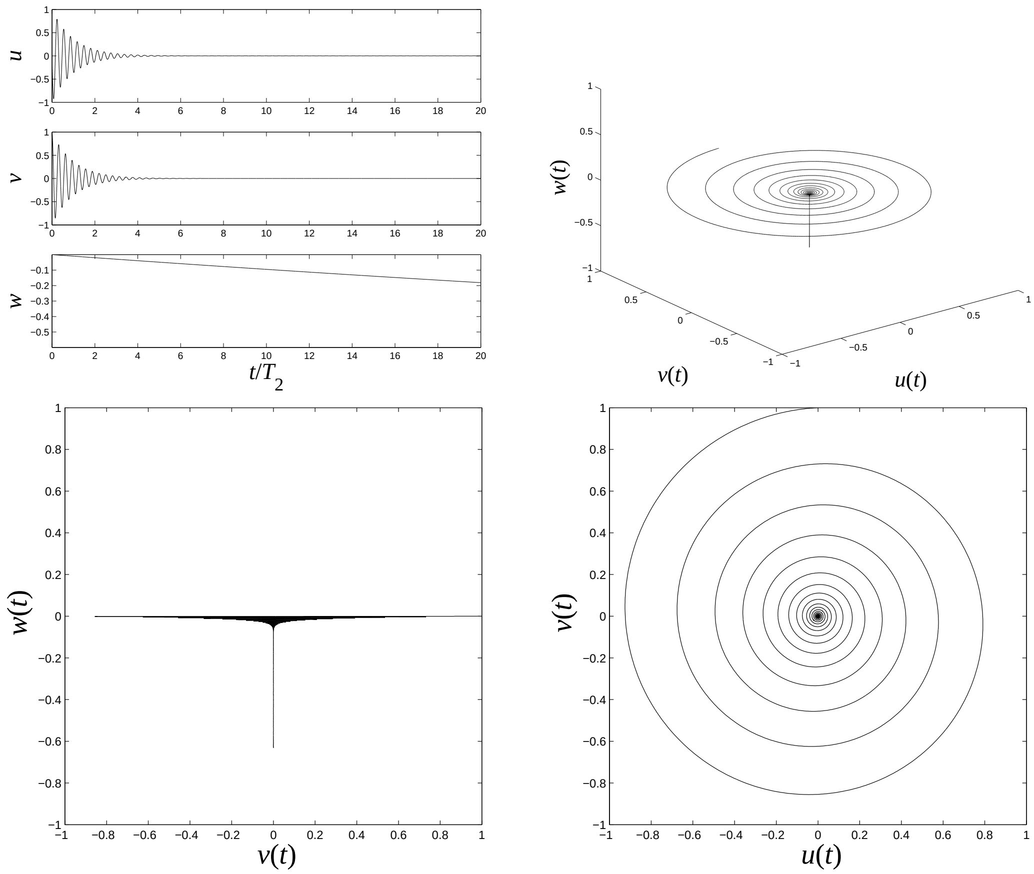


Figure 9a. Evolution of the Bloch vector $(u(t), v(t), w(t))$ after the optical field is switched off, for the far off-resonant case ($\delta \neq 0$), and with the longitudinal relaxation time being much greater than the transverse relaxation time ($T_1 \gg T_2$). The parameters used in the simulation are $\eta \equiv T_1/T_2 = 100$, $\delta \equiv \Delta T_2 = 20$, $w_0 = -1$, and $\gamma(t) \equiv \beta(t)T_2 = 0$. (Compare with Fig. 5 for the exactly resonant case, and with Fig. 7a for the slightly off-resonant case.)

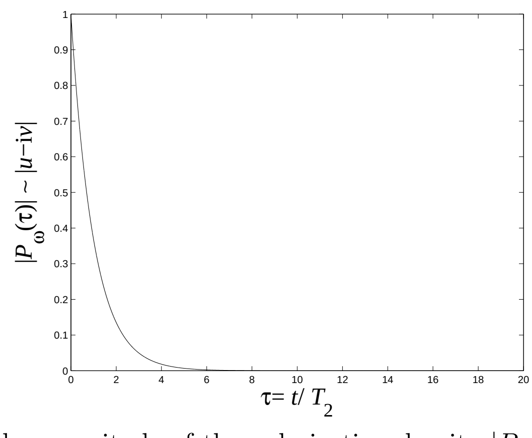


Figure 9b. Evolution of the magnitude of the polarization density $|P_\omega(t)| \sim |u(t) - iv(t)|$ as the optical field is switched on, corresponding to the simulation shown in Fig. 9a.

*The case $T_1 \approx T_2$ – Longitudinal relaxation approximately equal to transverse relaxation*

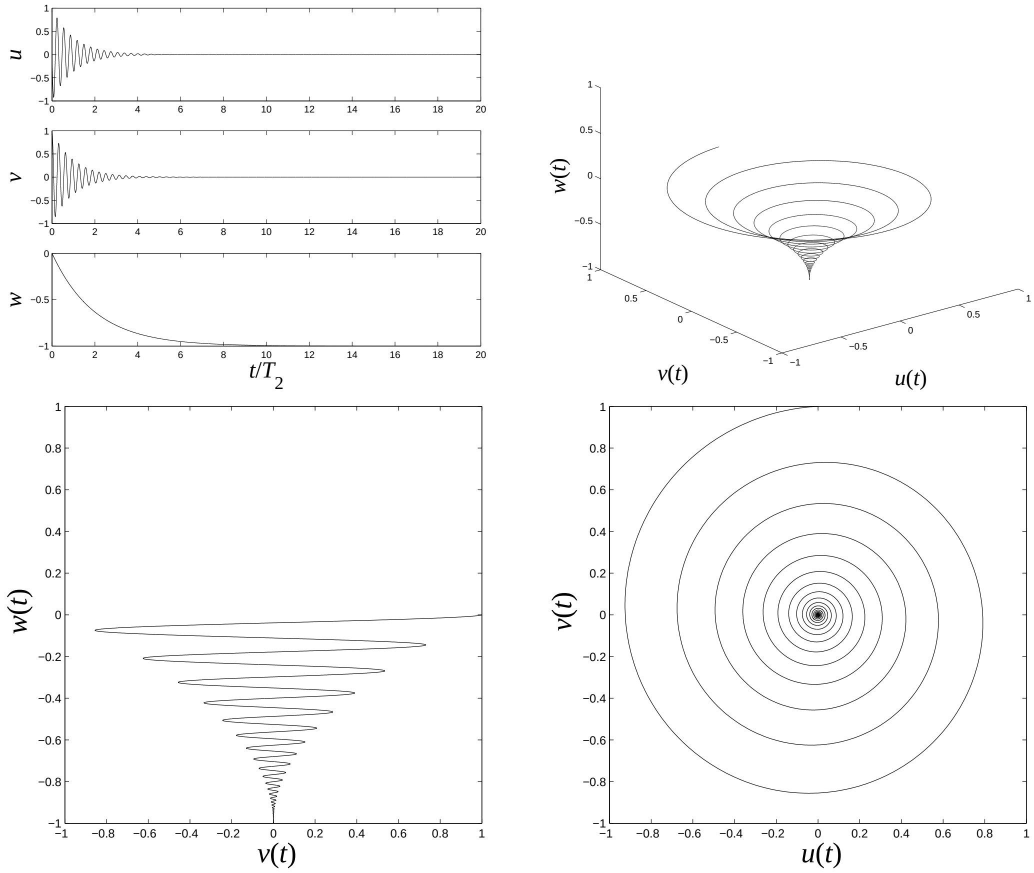


Figure 10a. Evolution of the Bloch vector $(u(t), v(t), w(t))$ after the optical field is switched off, for the far off-resonant case ($\delta \neq 0$), and with the longitudinal relaxation time being approximately equal to the transverse relaxation time ($T_1 \approx T_2$). The parameters used in the simulation are $\eta \equiv T_1/T_2 = 2$, $\delta \equiv \Delta T_2 = 20$, $w_0 = -1$, and $\gamma(t) \equiv \beta(t)T_2 = 0$. (Compare with Fig. 6 for the exactly resonant case, and with Fig. 8a for the slightly off-resonant case.)

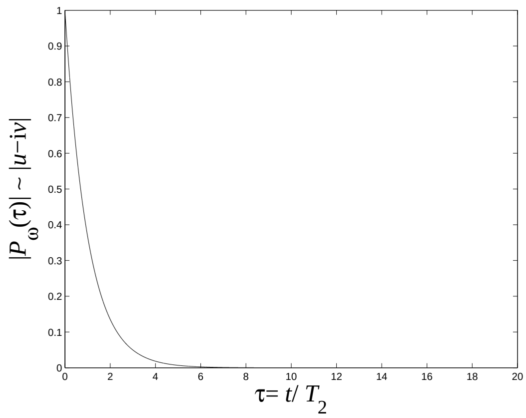


Figure 10b. Evolution of the magnitude of the polarization density $|P_\omega(t)| \sim |u(t) - iv(t)|$ as the optical field is switched on, corresponding to the simulation shown in Fig. 10a.

*The case $T_1 \ll T_2$ – Longitudinal relaxation faster than transverse relaxation*

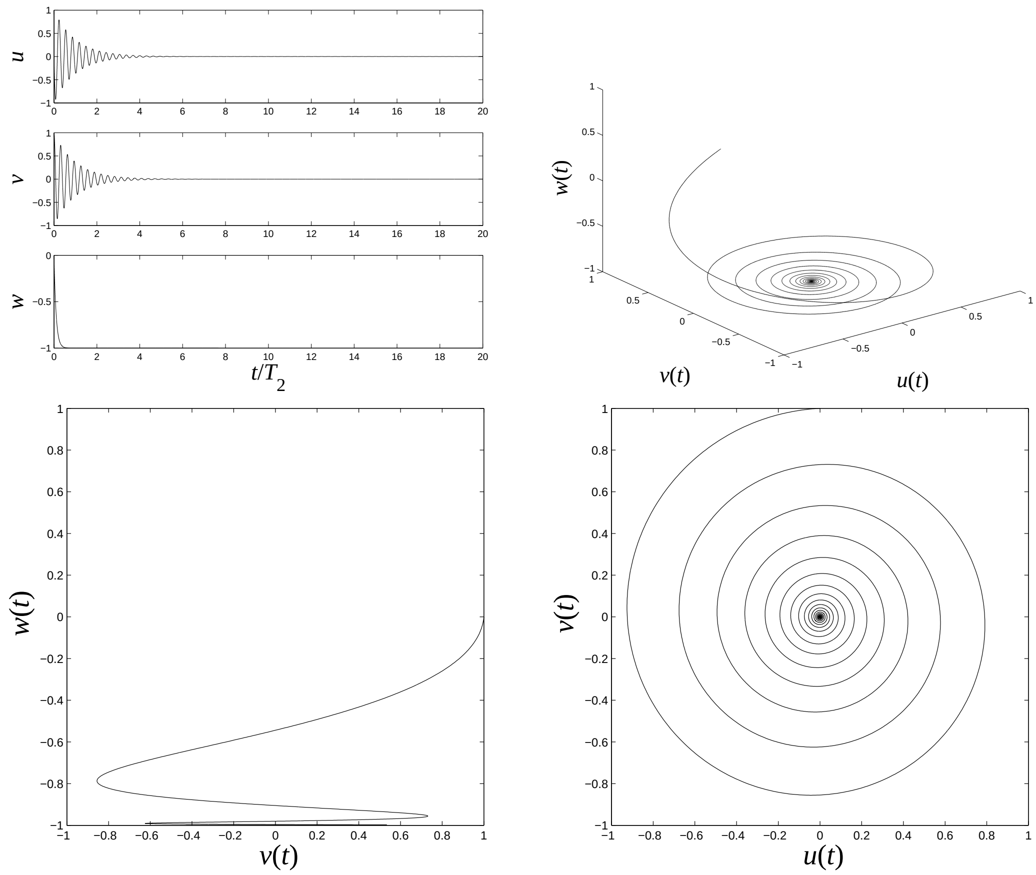


Figure 11a. Same parameter values as in Fig. 6, but with the longitudinal relaxation time being much smaller than the transverse relaxation time ($T_1 \ll T_2$), $\eta \equiv T_1/T_2 = 0.1$. (Compare with Figs. 9a and 10a for the cases $T_1 \gg T_2$ and $T_1 \approx T_2$, respectively.)

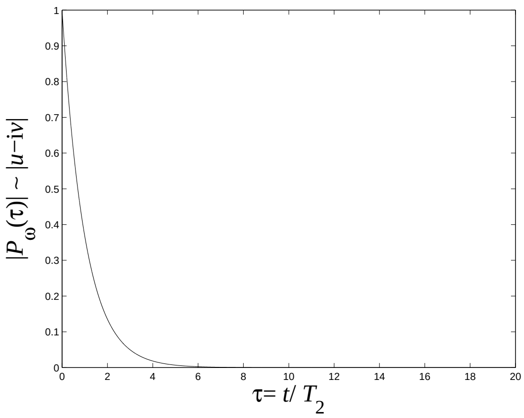


Figure 11b. Evolution of the magnitude of the polarization density $|P_\omega(t)| \sim |u(t) - iv(t)|$ as the optical field is switched on, corresponding to the simulation shown in Fig. 11a.

**The connection between the Bloch equations and the susceptibility**

As an example of the connection between the polarization density obtained from the Bloch equations and the one obtained from the susceptibility formalism, we will now – once again – consider the intensity-dependent refractive of the medium.

*The intensity-dependent refractive index in the susceptibility formalism*

Previously in this course, the intensity-dependent refractive index has been obtained from the optical Kerr-effect in isotropic media, in the form

$$n = n_0 + n_2|\mathbf{E}_\omega|^2,$$

where $n_0 = [1 + \chi^{(1)}_{xx}(-\omega;\omega)]^{1/2}$ is the linear refractive index, and

$$n_2 = \frac{3}{8n_0}\chi^{(3)}_{xxxx}(-\omega;\omega,\omega,-\omega)$$

is the parameter of the intensity dependent contribution. However, since we by now are fully aware that the polarization density in the description of the susceptibility formalism originally is given as an infinity series expansion, we may expect that the general form of the intensity dependent refractive index rather would be as a power series in the intensity,

$$n = n_0 + n_2|\mathbf{E}_\omega|^2 + n_4|\mathbf{E}_\omega|^4 + n_6|\mathbf{E}_\omega|^6 + \ldots$$

For linearly polarized light, say along the $x$-axis of a Cartesian coordinate system, we know that such a series is readily possible to derive in terms of the susceptibility formalism, with the different order terms of the refractive index expansion given by the elements

$$\begin{aligned}
n_2 &\sim \chi^{(3)}_{xxxx}(-\omega;\omega,\omega,-\omega),\\
n_4 &\sim \chi^{(5)}_{xxxxxx}(-\omega;\omega,\omega,-\omega,\omega,-\omega),\\
n_6 &\sim \chi^{(7)}_{xxxxxxxx}(-\omega;\omega,\omega,-\omega,\omega,-\omega,\omega,-\omega),\\
&\vdots
\end{aligned}$$

Such an analysis would, however, be extremely cumbersome when it comes to the analysis of higher-order effects, and the obtained sum of various order terms would also be almost impossible to obtain a closed expression for. For future reference, to be used in the interpretation of the polarization density given by the Bloch equations, the intensity dependent polarization density is though shown in its explicit form below, including up to the seventh order interaction term in the Butcher and Cotter convention,

$$\begin{aligned}
P^x_\omega = \varepsilon_0\chi^{(1)}_{xx}(-\omega;\omega)E^x_\omega & \qquad \text{(order } n=1)\\
+ \varepsilon_0(3/4)\chi^{(3)}_{xxxx}(-\omega;\omega,\omega,-\omega)|E^x_\omega|^2E^x_\omega & \qquad \text{(order } n=3)\\
+ \varepsilon_0(5/8)\chi^{(5)}_{xxxxxx}(-\omega;\omega,\omega,-\omega,\omega,-\omega)|E^x_\omega|^4E^x_\omega & \qquad \text{(order } n=5)\\
+ \varepsilon_0(35/64)\chi^{(7)}_{xxxxxxxx}(-\omega;\omega,\omega,-\omega,\omega,-\omega,\omega,-\omega)|E^x_\omega|^6E^x_\omega & \qquad \text{(order } n=7)\\
+ \ldots &
\end{aligned}$$

The other approach to calculation of the polarization density, as we next will outline, is to use the steady-state solutions to the Bloch equations.

*The intensity-dependent refractive index in the Bloch-vector formalism*

For steady-state interaction between light and matter, the solutions to the Bloch equations yield

$$u - iv = \frac{-\beta w}{\Delta - i/T_2}, \qquad \text{[B. \& C. (6.53a)]},$$

$$w = \frac{w_0[1 + (\Delta T_2)^2]}{1 + (\Delta T_2)^2 + \beta^2 T_1 T_2}, \qquad \text{[B. \& C. (6.53b)]},$$

where, as previously, $\beta = er^{\alpha}_{ab}E^{\alpha}_{\omega}(t)/\hbar$ is the Rabi frequency, though now considered to be a slowly varying (adiabatically following) quantity, due to the assumption of steady-state behaviour. From the steady-state solutions, the $\mu$-component ($\mu = x, y, z$) of the electric polarization density $\mathbf{P}(\mathbf{r}, t) = \mathrm{Re}[\mathbf{P}_\omega \exp(-i\omega t)]$ of the medium hence is given as

$$\begin{aligned}
P^{\mu}_{\omega} &= Ner^{\mu}_{ab}(u - iv)\\
&= -Ner^{\mu}_{ab}\frac{\beta w}{\Delta - i/T_2}\\
&= -Ner^{\mu}_{ab}\frac{\beta}{(\Delta - i/T_2)}\frac{w_0[1 + (\Delta T_2)^2]}{[1 + (\Delta T_2)^2 + \beta^2 T_1 T_2]}\\
&= -New_0\frac{r^{\mu}_{ab}}{(\Delta - i/T_2)}\frac{\beta}{\left[1 + \frac{T_1 T_2}{(1+(\Delta T_2)^2)}\beta^2\right]}.
\end{aligned} \tag{3}$$

In this expression for the polarization density, it might at a first glance seem as it is negative for a positive Rabi frequency $\beta$, henc giving a polarization density that is directed anti-parallel to the electric field. However, the quantity $w_0 = \rho_0(b) - \rho_0(a)$, the population inversion at thermal equilibrium, is always negative (since we for sure do not have any population inversion at thermal equilibrium, for which we rather expect the molecules to occupy the lower state), hence ensuring that the off-resonant, real-valued polarization density always is directed along the direction of the electric field of the light.

Next observation is that the polarization density no longer is expressed as a power series in terms of the electric field, but rather as a rational function,

$$P^{\mu}_{\omega} \sim X/(1 + X^2), \tag{4}$$

where

$$\begin{aligned}
X &= \sqrt{T_1 T_2/(1 + (\Delta T_2)^2)}\beta\\
&= \sqrt{T_1 T_2/(1 + (\Delta T_2)^2)}er^{\alpha}_{ab}E^{\alpha}_{\omega}(t)/\hbar
\end{aligned}$$

is a parameter linear in the electric field. The principal shape of the rational function in Eq. (4) is shown in Fig. 12.

From Eq. (4), the polarization density is found to increase with increasing $X$ up to $X = 1$, as we expect for an increasing power of an optical beam. However, for $X > 1$, we find the somewhat surprising fact that the polarization density instead *decrease* with an increasing intensity; this peculiar suggested behaviour should hence be explained before continuing.

The first observation we may do is that the linear polarizability (i. e. what we usually associate with linear optics) follows the first order approximation $p(X) = X$. In the region where the peculiar decrease of the polarization density appear, the difference between the suggested nonlinear polarization density and the one given by the linear approximation is *huge*, and since we *a priori* expect nonlinear contributions to be small compared to the alsways present linear ones, this is already an indication of that we in all practical situations do not have to consider the descrease of polarization density as shown in Fig. 12.

For optical fields of the strength that would give rise to nonlinearities exceeding the linear terms, the underlying physics will rather belong to the field of plasma and high-energy physics, rather than a bound-charge description of gases and solids. This implies that the validity of the models here applied (bound charges, Hamiltonians being linear in the optical field, etc.) are limited to a range well within $X \leq 1$.

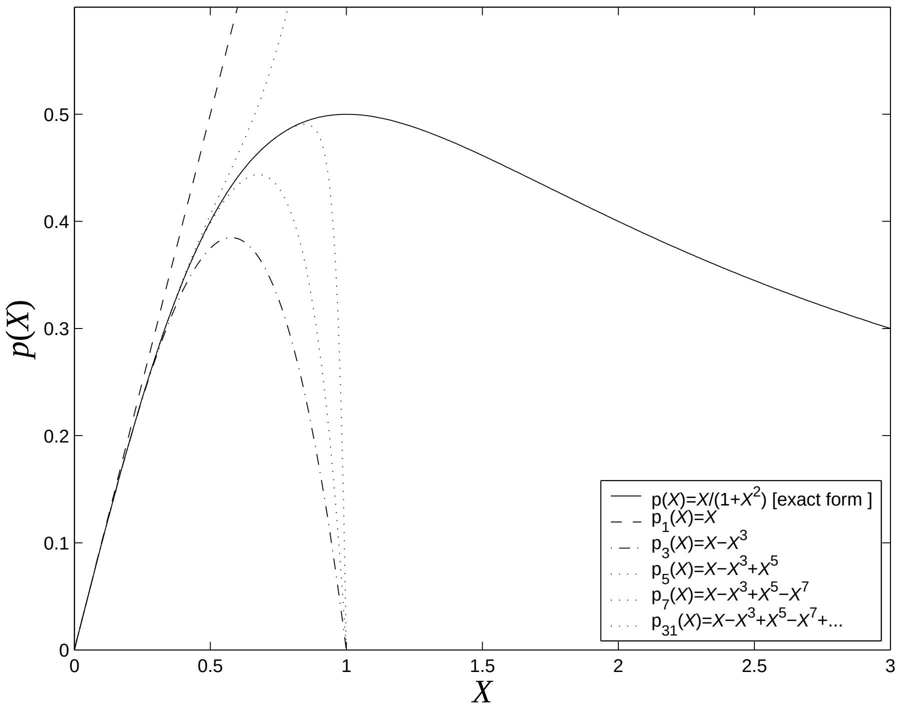


Figure 12. The principal shape of the electric polarization density of the medium, as function of the applied electric field of the light. In this figure, $X = \sqrt{T_1T_2/(1+(\Delta T_2)^2)}\beta$ is a normalized parameter describing the field strength of the electric field of the light.

Another interesting point we may observe is a more mathematically related one. In Fig. 12, we see that even for very high order terms (such as the approximating power series of degree 31, as shown in the figure), all power series expansions fail before reaching $X = 1$. The reason for this is that the power series that approximate the rational function $X/(1 + X^2)$,

$$X/(1+X^2) = X - X^3 + X^5 - X^7 + \ldots,$$

is convergent only for $|X| < 1$; for all other values, the series is divergent. This means that no matter how many terms we include in the power series in $X$, it will nevertheless fail when it comes to the evaluation for $|X| > 1$. Since this power series expansion is equivalent to the expansion of the nonlinear polarization density in terms of the electrical field of the light (keeping in mind that $X$ here actually is linear in the electric field and hence strictly can be considered as the field variable), this also is an indication that at this point the whole susceptibility formalism fail to give a proper description at this working point.

This is an excellent illustration of the downturn of the susceptibility description of interaction between light and matter; no matter how many terms we may include in the power series of the electrcal field, *it will at some point nevertheless fail to give the total picture of the interaction*, and we must then instead seek other tools.

Returning to the polarization density given by Eq. (3), we may now express this in an explicit form by inserting $\Delta \equiv \Omega_{ba} - \omega$ for the angular frequency detuning, the Rabi frequency $\beta = er^\alpha_{ab}E^\alpha_\omega(t)/\hbar$, and the thermal equilibrium inversion $w_0 = \rho_0(b) - \rho_0(a)$. This gives the polarization density of the medium as

$$P^\mu_\omega = \varepsilon_0 \underbrace{\underbrace{\frac{Ne^2}{\varepsilon_0\hbar}(\rho_0(a)-\rho_0(b))\frac{r^\mu_{ab}r^\alpha_{ab}}{(\Omega_{ba}-\omega-i/T_2)}}_{=\chi^{(1)}_{\mu\alpha}(-\omega;\omega)\text{ for a two level medium}} \underbrace{\frac{1}{\left[1+\frac{T_1T_2}{(1+(\Omega_{ba}-\omega)^2T_2^2)}(er^\gamma_{ab}E^\gamma_\omega/\hbar)^2\right]}}_{\text{nonlinear correction factor to }\chi^{(1)}_{\mu\alpha}(-\omega;\omega)}}_{\text{the field corrected susceptibility, }\bar{\chi}(\omega;\mathbf{E}_\omega)\text{ [see Butcher and Cotter, section 6.3.1]}} E^\alpha_\omega.$$

In this form, the polarization density is given as the product with a term which is identical to the linear susceptibility[4] (as obtained in the perturbation analysis in the frame of the susceptibility formalism), and a correction factor which is a nonlinear function of the electric field.

The nonlinear correction factor, of the form $1/(1+X^2)$, with $X = \sqrt{T_1T_2/(1+(\Delta T_2)^2)}\beta$ as previously, can now be expanded in a power series around the small-signal limit $X = 0$, using

$$1/(1+X^2) = 1 - X^2 + X^4 - X^6 + \dots,$$

from which we obtain the polarization density as a power series in the electric field (which for the sake of simplicitly now is taken as linearly polarized along the $x$-axis) as

$$\begin{aligned}
P^x_\omega \approx\ & \varepsilon_0 \frac{Ne^2}{\varepsilon_0\hbar}(\rho_0(a)-\rho_0(b))\frac{r^x_{ab}r^x_{ab}}{(\Omega_{ba}-\omega-i/T_2)}E^x_\omega \\
& - \varepsilon_0\frac{Ne^4}{\varepsilon_0\hbar^3}(\rho_0(a)-\rho_0(b))\frac{r^x_{ab}r^x_{ab}}{(\Omega_{ba}-\omega-i/T_2)}\frac{(r^x_{ab})^2}{[1/T_2^2+(\Omega_{ba}-\omega)^2](T_2/T_1)}|E^x_\omega|^2E^x_\omega \\
& + \varepsilon_0\frac{Ne^6}{\varepsilon_0\hbar^5}(\rho_0(a)-\rho_0(b))\frac{r^x_{ab}r^x_{ab}}{(\Omega_{ba}-\omega-i/T_2)}\frac{(r^x_{ab})^4}{[1/T_2^2+(\Omega_{ba}-\omega)^2]^2(T_2/T_1)^2}|E^x_\omega|^4E^x_\omega \\
& + \dots
\end{aligned} \tag{5}$$

This form is identical to one as obtained in the susceptibility formalism; however, the steps that led us to this expression for the polarization density *do not rely on the perturbation theory of the density operator*, but rather on the explicit form of the steady-state solutions to the Bloch equations.

**Summary of the Bloch and susceptibility polarization densities**

To summarize this last lecture on the Bloch equations, expressing the involved parameters in the same style as previously used in the description of the susceptibility formalism, the polarization density obtained from the steady-state solutions to the Bloch equations is

$$P^\mu_\omega = \varepsilon_0\frac{Ne^2}{\varepsilon_0\hbar}(\rho_0(a)-\rho_0(b))\frac{r^\mu_{ab}r^\alpha_{ab}}{(\Omega_{ba}-\omega-i/T_2)}\frac{1}{\left[1+\frac{T_1T_2}{(1+(\Omega_{ba}-\omega)^2T_2^2)}(er^\alpha_{ab}E^\alpha_\omega/\hbar)^2\right]}E^\alpha_\omega.$$

By expanding this in a power series in the electrical field, one obtains the form (5), in which we from the same description of the polarization density in the susceptibility formalism can identify

$$\begin{aligned}
\chi^{(1)}_{xx}(-\omega;\omega) &= \frac{Ne^2}{\varepsilon_0\hbar}(\rho_0(a)-\rho_0(b))\frac{r^x_{ab}r^x_{ab}}{(\Omega_{ba}-\omega-i/T_2)}, \\
\chi^{(3)}_{xxxx}(-\omega;\omega,\omega,-\omega) &= -\frac{4Ne^4}{3\varepsilon_0\hbar^3}(\rho_0(a)-\rho_0(b))\frac{r^x_{ab}r^x_{ab}}{(\Omega_{ba}-\omega-i/T_2)}\frac{(r^x_{ab})^2}{[1/T_2^2+(\Omega_{ba}-\omega)^2](T_2/T_1)}, \\
\chi^{(5)}_{xxxxxx}&(-\omega;\omega,\omega,-\omega,\omega,-\omega) \\
&= \frac{8Ne^6}{5\varepsilon_0\hbar^5}(\rho_0(a)-\rho_0(b))\frac{r^x_{ab}r^x_{ab}}{(\Omega_{ba}-\omega-i/T_2)}\frac{(r^x_{ab})^4}{[1/T_2^2+(\Omega_{ba}-\omega)^2]^2(T_2/T_1)^2},
\end{aligned}$$

as being the first contributions to the two-level polarization density, including up to fifth order interactions. For a summary of the non-resonant forms of the susceptibilities of two-level systems, se Butcher and Cotter, Eqs. (6.71)–(6.73).

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[4] In the explicit expressions for the linear susceptibility, for example Butcher and Cotter's Eqs. (4.58) and (4.111) for the non-resonant and resonant cases, respectively, there are two terms, one with $\Omega_{ba}-\omega$ in the denominator and the other one with $\Omega_{ba}+\omega$. The reason why the second form does not appear in the expression for the field corrected susceptibility, as derived from the Bloch equations, is that *we have used the rotating wave approximation in the derivation of the final expression.* (Recapitulate that in the rotating wave approximation, terms with oscillatory dependence of $\exp[i(\Omega_{ba}+\omega)t]$ were neglected.) As a result, all temporally phase-mismatched terms are neglected, and in particular only terms with $\Omega_{ba}-\omega$ in the denominator will remain. This, however, is a most acceptable approximation, especially when it comes to resonant interactions, where terms with $\Omega_{ba}-\omega$ in the denominator by far will dominate over non-resonant terms.

**Appendix: Notes on the numerical solution to the Bloch equations**

In their original form, the Bloch equations for a two-level system are given by Eqs. (1) as

$$\begin{aligned} \frac{du}{dt} &= -\Delta v - u/T_2, \\ \frac{dv}{dt} &= \Delta u + \beta(t)w - v/T_2, \\ \frac{dw}{dt} &= -\beta(t)v - (w - w_0)/T_1. \end{aligned}$$

By taking the time in units of the transverse relaxation time $T_2$, as

$$\tau = t/T_2,$$

the Bloch equations in this normalized time scale become

$$\begin{aligned} \frac{du}{d\tau} &= -\Delta T_2 v - u, \\ \frac{dv}{d\tau} &= \Delta T_2 u + \beta(t)T_2 w - v, \\ \frac{dw}{d\tau} &= -\beta(t)T_2 v - (w - w_0)T_2/T_1. \end{aligned}$$

In this system of equations, all coefficients are now normalized and physically dimensionless, expressed as relevant quotes between relaxation times and products of the Rabi frequency or detuning frequency with the transverse relaxation time. Hence, by taking the normalized parameters

$$\begin{aligned} \delta &= \Delta T_2, \\ \gamma(t) &= \beta(t)T_2, \\ \eta &= T_1/T_2, \end{aligned}$$

where $\delta$ can be considered as the normalized detuning from molecular resonance of the medium, $\gamma(t)$ as the normalized Rabi frequency, and $\eta$ as a parameter which describes the relative impact of the longitudinal vs transverse relaxation times, the Bloch equations take the normalized final form

$$\begin{aligned} \frac{du}{d\tau} &= -\delta v - u, \\ \frac{dv}{d\tau} &= \delta u + \gamma(t)w - v, \\ \frac{dw}{d\tau} &= -\gamma(t)v - (w - w_0)/\eta. \end{aligned}$$

This normalized form of the Bloch equations has been used throughout the generation of graphs in Figs. 2–11 of this lecture, describing the qualitative impact of different regimes of resonance and relaxation. The normalized Bloch equations were in the simulations shown in Figs. 2–11 integrated by using the standard routine `ODE45()` in MATLAB.