Dynamics of a two-level atom in a broadband light field

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We provide a fully analytical description of a two-level atom interacting with a broadband light field. The problem we present is a typical example of a physical situation which occurs very commonly in practice, but is not straightforwardly solvable. However, we show in this paper that, remarkably, only a limited number of elementary calculations are required to treat the problem. The presented results are not only highly intuitive, but also allow us to study the nontrivial nonlinear response of a two-level atom to incident broadband radiation.

I. INTRODUCTION

The interaction of a two-level atom and a monochromatic field has always been a very popular topic in quantum physics. The attraction of this system is due to the relatively modest mathematical tools needed to describe the problem, combined with a rich physical behavior, able to accurately predict many interesting phenomena such as superradiance or the Mollow triplet.

The effect of the incident field on the atom's dynamics is described by the optical Bloch equations. This set of equations allows one to obtain expressions for the average atomic level populations and coherences in the presence of an incident field. If the incident light is monochromatic, the optical Bloch equations are easily dealt with. For broadband incident light, however, solving the optical Bloch equations is in general a far from trivial but physically very relevant matter. For example, a common experimental situation we are considering is an atom or molecule probed by near-resonant light, but at the same time irradiated by a broadband light source, such as a trapping laser.

In this paper, we show how to transparently describe the interaction of a two-level system and a broadband incident wave. An everyday (and practically important) example of a broadband field is the field of spontaneous photons emitted by an atom. The spectrum corresponding to spontaneous emission has a Lorentzian frequency distribution, and is therefore broadband. Obviously, we intuitively expect that if the spectrum of the incident field is far detuned compared to the resonance frequency of the atom, or very broad compared to the atom's natural linewidth, no significant interaction will take place. Likewise, if the incident field has a very narrow frequency distribution, we expect that the interaction will resemble the one induced by a monochromatic incident wave. In this paper, we show that only a few elementary calculations are required to quantify the interaction of such a broadband field and a two-level atom. We find that not just the incident spectrum itself, but in addition the overlap of the incident spectrum and the natural Lorentzian emission line of the atom itself determines the interaction strength, confirming our intuitive predictions.

In order to simplify the calculations, we will impose a very general restriction on the incident field. More precisely, we consider in this paper the class of statistically stationary incident fields. The mathematical simplifications they allow for, and the fact that many fields encountered in practice are statistically stationary, is the reason why they are treated in so many textbooks on, e.g., quantum optics or magnetic resonance. An important property of such fields is that two-time averages of $\langle E(t)E(t')\rangle$ of the field $E(t)$ only depend on the time difference $t-t'$, which implies that field components at different frequencies are uncorrelated.

Finally, we will apply a standard approximation to the evolution of the atom itself: we will average out the contribution of highly non-resonating terms. For a monochromatic incident wave, for example, this approximation simply implies that all components oscillating at twice the incident wave frequency are neglected. The physical meaning of this approximation is that non-energy conserving emission and absorption of photons is neglected, or in other words, that “virtual photons” are not taken into account. For a monochromatic incident wave, this approximation is referred to in the literature as the Rotating Wave Approximation (RWA). Since we deal in this paper with fields which are in general broadband, we will extend the standard approximation found in textbooks, and refer to the extension as the Generalized Rotating Wave Approximation (GRWA).

In the following, we will start by formulating the optical Bloch equations for our configuration. A Fourier transform will lead to a better understanding of the behavior of our system in frequency space. Application of the GRWA will return steady-state solutions for the optical Bloch equations.

To conclude our paper, we will show that the results obtained can be straightforwardly used to express the nonlinear response of an atom to a broadband field. This response, which can be quantified by the atom’s dynamic polarizability, is of primary importance in studying atom-field interactions, since it allows one to gain full insight into the internal atomic dynamics.

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II. THE OPTICAL BLOCH EQUATIONS

We consider a two-level atom $A$ with lower level $a$ and upper level $c$, separated by an energy difference $\hbar \omega_{ca}$. The atom interacts with an incident time-dependent real-valued field $E(t)$. The dynamics of the system can be described in terms of the density matrix $\hat{\sigma}(t)$. The time evolution of $\hat{\sigma}(t)$ is given by the optical Bloch equations, which can be written, in the electric dipole approximation, as

\begin{align}
\dot{\sigma}_{cc} &= +i \Omega(t) \left( \sigma_{ca} - \sigma_{ac} \right) - \Gamma \sigma_{cc}, \\
\dot{\sigma}_{aa} &= -i \Omega(t) \left( \sigma_{ca} - \sigma_{ac} \right) + \Gamma \sigma_{cc}, \\
\dot{\sigma}_{ac} &= -i \Omega(t) \left( \sigma_{cc} - \sigma_{aa} \right) + i \omega_{ca} \sigma_{ac} - \frac{\Gamma}{2} \sigma_{ac}, \\
\dot{\sigma}_{ca} &= +i \Omega(t) \left( \sigma_{cc} - \sigma_{aa} \right) - i \omega_{ca} \sigma_{ca} - \frac{\Gamma}{2} \sigma_{ca},
\end{align}

where the notation $\dot{f}(t) \equiv \frac{d}{dt} f(t)$ is used. Equations (1) can be found in many elementary books on quantum optics [12]. The matrix elements $\sigma_{aa}$ and $\sigma_{cc}$ are the ensemble-averaged populations of the lower and upper atomic level, respectively. They are related by $\sigma_{aa} + \sigma_{cc} = 1$, expressing conservation of population. The off-diagonal elements represent coherences; we will elaborate on their physical meaning in section IV. The constant decay rate $\Gamma$ represents spontaneous emission by the system to the surrounding vacuum. The Rabi frequency $\Omega(t) \equiv -\frac{1}{\hbar} d_{ac} \cdot E(t)$ quantifies the interaction strength between the atom and the incident field, with $d_{ac}$ the $c \rightarrow a$ transition dipole moment. In the conventional case of a monochromatic incident wave $E(t) \equiv E_0 \cos \omega_L t$ which is often found in the literature, the definition $\Omega_{\text{Rabi}} \equiv -\frac{1}{\hbar} d_{ac} \cdot E_0$ is mostly used, explicitly removing the oscillatory time dependence of the field from the Rabi frequency (obviously, the optical Bloch equations then contain extra factors $e^{\pm i \omega_L t}$). However, we will see that for a more general time dependence, as the one we deal with here, it is beneficial to use our definition and consider a time-dependent Rabi frequency.

Our aim in this section of the paper is to derive steady-state solutions for equations (1). The statistical properties of the field are especially advantageous in the frequency domain, since if the two-time average $\langle \Omega(t) \Omega(t + \tau) \rangle$ of the phase of $\Omega$ only depends on $\tau$, one can deduce (see, e.g., [13]) that in the frequency domain

$$
\langle \Omega(\omega) \Omega(\omega') \rangle = J[\omega] \delta(\omega + \omega'),
$$

where $J[\omega]$ is the spectral density function of the incident radiation and the Fourier transform of $\Omega(t)$ is defined as

$$
\Omega [\omega] \equiv \frac{1}{2\pi} \int_{-\infty}^{+\infty} \Omega(t) e^{-i\omega t} dt,
$$

$$
\Omega(t) = \int_{-\infty}^{+\infty} \Omega [\omega] e^{i\omega t} d\omega.
$$

We will see that the appearance of a delta function in (2) will facilitate the calculations further on. In what follows, we will Fourier transform the optical Bloch equations. We therefore define the population difference $w(t)$ and its Fourier transform $w[\omega]$ as

$$
w(t) \equiv \sigma_{cc}(t) - \sigma_{aa}(t) \equiv \int_{-\infty}^{+\infty} w[\omega] e^{-i\omega t} dt,
$$

and

$$
\sigma_{ac}(t) \equiv \int_{-\infty}^{+\infty} \sigma_{ac}[\omega] e^{i\omega t} d\omega \equiv \int_{0}^{+\infty} \sigma_{ac}[\omega] e^{i\omega t} d\omega,
$$

$$
\sigma_{ca}(t) \equiv \int_{-\infty}^{+\infty} \sigma_{ca}[\omega] e^{i\omega t} d\omega \equiv \int_{-\infty}^{0} \sigma_{ca}[\omega] e^{i\omega t} d\omega,
$$

where the contribution of the non-resonant part of the coherences has been omitted. The restriction of the integration interval in expressions (5) has the simple physical meaning that non-energy conserving terms are not taken into account, as mentioned in the introduction of our paper. This approximation is a straightforward generalization of the Rotating Wave Approximation [14] and is therefore referred to as the Generalized Rotating Wave Approximation. This generalization holds excellently if $J[\omega]$ is only appreciably different from zero near the atomic resonance.

If we now split the Fourier transform $\Omega[\omega]$ of the Rabi frequency $\Omega(t)$ into a positive- and a negative-frequency part

$$
\Omega(t) = \int_{0}^{+\infty} \Omega [\omega] e^{i\omega t} d\omega + \int_{-\infty}^{0} \Omega [\omega] e^{i\omega t} d\omega
$$

$$
\equiv \Omega_+(t) + \Omega_-(t),
$$

we see that neglecting all highly non-resonant terms in (1) results in slightly altered optical Bloch equations:

\begin{align}
\dot{\sigma}_{cc} &= +i \Omega_+(t) \sigma_{ca} - i \Omega_-(t) \sigma_{ac} - \Gamma \sigma_{cc}, \\
\dot{\sigma}_{aa} &= -i \Omega_+(t) \sigma_{ca} + i \Omega_-(t) \sigma_{ac} + \Gamma \sigma_{cc}, \\
\dot{\sigma}_{ac} &= -i \Omega_+(t) \left( \sigma_{cc} - \sigma_{aa} \right) + i \omega_{ca} \sigma_{ac} - \frac{\Gamma}{2} \sigma_{ac}, \\
\dot{\sigma}_{ca} &= +i \Omega_-(t) \left( \sigma_{cc} - \sigma_{aa} \right) - i \omega_{ca} \sigma_{ca} - \frac{\Gamma}{2} \sigma_{ca}.
\end{align}
It is clear that equations (7) are far simpler to deal with than the original Bloch equations (1). Fourier transforming equation (6a) and (6b) gives

\[
(\Gamma + i\omega)w[\omega] = +2i \int_{0}^{+\infty} \Omega[\omega']\sigma_{ca}[\omega - \omega']d\omega' - 2i \int_{-\infty}^{0} \Omega[\omega']\sigma_{ac}[\omega - \omega']d\omega' - \Gamma \delta(\omega). \tag{8}
\]

Fourier transforming (7c) and (7d), on the other hand,

\[
\text{transforming equation (7a) and (7b) gives}
\]

\[
(w - \omega_{ca} + \frac{\Gamma}{2})\sigma_{ac}[\omega] = -i \int_{0}^{+\infty} \Omega[\omega'']w[\omega - \omega'']d\omega'', \tag{9a}
\]

\[
(i\omega + i\omega_{ca} + \frac{\Gamma}{2})\sigma_{ca}[\omega] = +i \int_{-\infty}^{0} \Omega[\omega'']w[\omega - \omega'']d\omega''. \tag{9b}
\]

If we now substitute the previous expressions in (8), we find

\[
(\Gamma + i\omega)w[\omega] + \Gamma \delta(\omega) =
\]

\[
= -2 \int_{0}^{+\infty} d\omega' \int_{-\infty}^{0} d\omega'' \Omega[\omega']\Omega[\omega'']w[\omega - \omega' - \omega''] \times \left( \frac{1}{i\omega - i\omega' + i\omega_{ca} + \frac{\Gamma}{2}} + \frac{1}{i\omega - i\omega'' - i\omega_{ca} + \frac{\Gamma}{2}} \right), \tag{10}
\]

which is a self-consistent equation in the population difference. We use the following trial solution for \(w[\omega]\):

\[
w[\omega] = w[0]\delta(\omega), \tag{11}
\]

which is appropriate since we are interested in the regime for which the populations are time-independent (which is also referred to as the “steady-state” regime). Substitution of (11) in (10) and phase-averaging yields

\[
\Gamma w[0] + \Gamma = -2w[0] \int_{0}^{+\infty} d\omega'J[\omega'] \left( \frac{1}{(\omega' - \omega_{ca})^2 + \left(\frac{\Gamma}{2}\right)^2} \right), \tag{12}
\]

and therefore

\[
w[\omega] = \frac{1}{1 + 2 \int_{0}^{+\infty} d\omega'J[\omega'] \left( \frac{1}{(\omega' - \omega_{ca})^2 + \left(\frac{\Gamma}{2}\right)^2} \right)} \delta(\omega). \tag{13}
\]

We can conclude that in steady-state, we find the desired solution

\[
\sigma_{ac}^{ss} = 1, \quad \sigma_{ca}^{ss} = \frac{X}{2X + 1}, \tag{14a}
\]

\[
\sigma_{ac}^{st} = (\sigma_{ac}^{st})* = \frac{1}{2X + 1} \int_{-\infty}^{0} \frac{i\Omega[\omega]}{-i\omega_{ca} - i\omega - \frac{\Gamma}{2}} e^{i\omega t} d\omega, \tag{14b}
\]

with

\[
X \equiv \int_{0}^{+\infty} d\omega J[\omega] \frac{1}{(\omega_{ca} - \omega)^2 + \left(\frac{\Gamma}{2}\right)^2}. \tag{15}
\]

Expressions (14) are the key result of our paper. The influence of the spectral properties of the incident field enters the dynamics of the density matrix through a single interaction parameter \(X\). The expression (15) for \(X\) is surprisingly simple and appealing: it is the overlap integral of the spectral density of the incident field, and the natural Lorentzian emission line of the two-level system itself. The structure of the interaction parameter confirms what we intuitively expect: resonant fields with a narrow distribution interact strongly with the atom, while the interaction with broad, far off-resonance fields is far less pronounced. As examples, we will in the next section focus on two specific and interesting values for \(X\).

\section{III. EXAMPLES}

As a first demonstration of equations (14), we will verify that the well-known expressions for an incident monochromatic field \(E(t) = E_{0} \cos(\omega_{L}t)\) with \(\omega_{L} > 0\). The corresponding Rabi frequency is

\[
\Omega[\omega] = \frac{\Omega_{0}}{2} \left( \delta(\omega - \omega_{L}) + \delta(\omega + \omega_{L}) \right), \tag{16}
\]

therefore

\[
\Omega[\omega]\Omega[\omega'] =
\]

\[
\frac{\Omega_{0}^2}{4} \left( \delta(\omega - \omega')\delta(\omega - \omega_{L}) + \delta(\omega - \omega')\delta(\omega + \omega_{L}) + \delta(\omega + \omega')\delta(\omega - \omega_{L}) + \delta(\omega + \omega')\delta(\omega + \omega_{L}) \right). \tag{17}
\]

Of the 4 terms appearing in (17), only the first remains in the (G)RWA, yielding

\[
J[\omega] = \frac{\Omega_{0}^2}{4} \delta(\omega - \omega_{L}). \tag{18}
\]
which transforms expressions (14) into

\[
\begin{align*}
\sigma_{st}^{st} &= \frac{(\Omega_0/2)^2}{(\omega_L - \omega_{ca})^2 + \left(\frac{\Gamma}{2}\right)^2} \frac{1}{\pi}, \\
\sigma_{st}^{0s} &= \frac{(\Omega_0/2)^2 e^{-\omega_L t}}{(\omega_L - \omega_{ca})^2 + \left(\frac{\Gamma}{2}\right)^2 + \frac{1}{2}(\omega_L - \omega_{ca})^2 - \frac{\Gamma}{2}}.
\end{align*}
\]

(19a)

(19b)

which corresponds exactly to the solutions for incident monochromatic fields found in the literature [12], justifying our method.

As a second demonstration of equations (14), we consider the nontrivial case of an atom interacting with incident spontaneous emission. The incident field then has a Lorentzian spectrum with a width \(\Gamma(1 + \kappa)\), \(\kappa \geq -1\), centered around \(\omega_L \gg \Gamma\):

\[
J[\omega] = \frac{J_0}{\pi} \frac{\Gamma}{(\omega - \omega_L)^2 + \left(\frac{\Gamma}{2}(1 + \kappa)\right)^2},
\]

(20)

where the factor

\[
J_0 = \int_{-\infty}^{\infty} J[\omega]d\omega = \langle \Omega(t)^2 \rangle
\]

(21)

is proportional to the total incident field energy. We find as an explicit solution for \(X\):

\[
X = \int_{0}^{\infty} d\omega J[\omega] \frac{1}{(\omega - \omega_{ca})^2 + \left(\frac{\Gamma}{2}\right)^2}
\]

\[
\approx \int_{-\infty}^{\infty} d\omega J[\omega] \frac{1}{(\omega - \omega_L)^2 + \left(\frac{\Gamma}{2}(1 + \kappa)\right)^2} \times \frac{1}{\pi} \frac{J_0\Gamma}{2(1 + \kappa)}
\]

\[
= J_0 \left(\frac{2 + \kappa}{(\omega_L - \omega_{ca})^2 + \left(\frac{\Gamma}{2}(2 + \kappa)\right)^2}\right),
\]

(22)

where the extension of the integral in (22) from 0 to \(-\infty\) is clearly justified by \(\omega_L \gg \Gamma\). Equation (22) clearly shows that the atom-field interaction is weak for spectrally broad or far-detuned fields, as mentioned earlier. To conclude this paper, we will in the next section use the result (14) with (19) and (22) obtained here to determine the nonlinear response of an atom to incident broadband light.

IV. APPLICATION

Let us now focus on a practical application of the previous sections. If one studies the response of an atom to incident light, one has to determine the strength with which the atom interacts with the incident light. A key property quantifying this strength is the atom’s dynamic polarizability \(\bar{\gamma}\). The associated polarization \(P\) induced by the incident field \(E_0\) is then given by \(P = \bar{\gamma} \cdot E_0\). In the limit of low-intensity incident fields, the response is linear and the polarization is then proportional to the incident field. However, at higher incident intensities, the response is no longer linear since the atom will start to saturate. The polarizability will then depend on the incident field and the polarization will no longer be proportional to the incident field. We will now show how the results from the previous sections can be straightforwardly used to quantify the rate at which the atom saturates due to a incident (broadband) field. In other words, we will show how expressions (14) allow us to calculate the nonlinear dynamic polarizability of a two-level atom.

We take the general case of an incident field \(E(t)\) consisting of a monochromatic cosine component with amplitude \(E_0\) and frequency \(\omega_L\), and a non-monochromatic component \(E_L(t)\), with a Lorentzian frequency distribution. The fields \(E(t)\) thus obtained define a very general class, containing the special cases of incident monochromatic light, as well as pure incident spontaneous emission. The Lorentzian component of the incident field has a linewidth \(\Gamma(1 + \kappa)\), \(\kappa \geq -1\), centered around \(\omega_L \gg \Gamma\):

\[
E[\omega] = E_0 \frac{1}{2} \left(\delta(\omega + \omega_L) + \delta(\omega - \omega_L)\right) + E_L[\omega].
\]

(23)

The corresponding Rabi frequencies are

\[
-h\Omega_0 = d_{ac} \cdot E_0,
\]

\[
-h\Omega_L[\omega] = d_{ac} \cdot E_L[\omega],
\]

(24a)

(24b)

obeying

\[
\langle \Omega_L[\omega]\Omega_L[\omega'] \rangle = J_L[\omega]\delta(\omega + \omega'),
\]

(25)

with \(J_L[\omega]\) the spectral density of the incident Lorentzian field

\[
J_L[\omega] = J_0 \frac{\Gamma}{\pi} \frac{\frac{\Gamma}{2}(1 + \kappa)}{(\omega - \omega_L)^2 + \left(\frac{\Gamma}{2}(1 + \kappa)\right)^2},
\]

(26)

Equations (23)-(26) fully describe the incident field. We will now focus on the response of the atom to this incident field. Since the dynamic polarizability and the density matrix both describe the response of the atom to incident light, both quantities should be connected.
Indeed, they are related by\(^1\)

\[
\varepsilon_0 \tilde{\alpha}(\omega) \cdot E[\omega] = d_{ac} \sigma_{ca}[\tilde{\omega}], \quad \omega > 0, \quad (27)
\]

Our general result\(^2\) now allows us to deduce that

\[
\varepsilon_0 \tilde{\alpha}(\omega) \cdot \left(\frac{1}{2} E_0 \delta(\omega + \omega_L) + E_L[\omega]\right) = \frac{d_{ac}}{2X+1} \left(\frac{1}{2} \Omega_0 \delta(\omega + \omega_L) + \Omega_L[\omega]\right),
\]

\[
= \frac{d_{ac}}{2X+1} \left(\frac{1}{2} \frac{\Omega_0 \omega_L}{\omega_L - \omega - i\frac{\Gamma}{2}} + \frac{\Omega_L[\omega]}{\omega_L - \omega - i\frac{\Gamma}{2}}\right),
\]

\[
= \frac{d_{ac}}{2X+1} \left(\frac{1}{2} \frac{\Omega_0 E_0}{\omega_L - \omega - i\frac{\Gamma}{2}} + \frac{d_{ac} \cdot E_L[\omega]}{\omega_L - \omega - i\frac{\Gamma}{2}}\right),
\]

(28)

where \(\otimes\) represents the tensor product of two vectors.

The nonlinear polarizability is thus expressed as

\[
\tilde{\alpha}(\omega) = \frac{d_{ac} \otimes d_{ac}}{\varepsilon_0 \hbar} \left[\frac{1}{2X+1} \left(\frac{1}{2} \frac{\Omega_0}{\omega_L - \omega - i\frac{\Gamma}{2}} + \frac{\Omega_L[\omega]}{\omega_L - \omega - i\frac{\Gamma}{2}}\right)\right],
\]

\[
= \frac{-\alpha_0}{2} \frac{1}{\omega_L - \omega - i\frac{\Gamma}{2}},
\]

(29)

where the static polarizability is given by

\[
\alpha_0 \equiv \frac{2}{\omega_{ca} \hbar} d_{ac} \otimes d_{ac},
\]

(30)

and where the saturation parameter can be written in a surprisingly simple way as

\[
S = \frac{\Omega_0^2}{4} + \int_0^{\infty} d\omega J_L[\omega] \left(\frac{(\omega_L - \omega_{ca})^2 + (\frac{\Gamma}{2})^2}{(\omega - \omega_{ca})^2 + (\frac{\Gamma}{2})^2}\right).
\]

(31)

Expressions\(^3\)–\(^5\) fully describe the response of a two-level atom to an incident field of the general class\(^6\).

Two limits for the incident field are in particular interesting. Firstly, for \(J_L[\omega] \to 0\), the expression for the dynamic polarizability of a two-level atom irradiated by a monochromatic field is recovered

\[
\tilde{\alpha}(\omega) = -\frac{1}{2} \frac{\omega_{ca}}{\omega - \omega_{ca} + i\frac{\Gamma}{2}},
\]

(32)

Secondly, for \(\Omega_0 \to 0\), we find the saturation due to pure spontaneous emission

\[
\tilde{\alpha}(\omega_L) = -\frac{1}{2} \frac{\omega_{ca}}{2(2X+1)} \frac{\omega_{ca}}{\omega_L - \omega_{ca} + i\frac{\Gamma}{2}},
\]

(33)

with \(X\) given by expression\(^7\).

Finally, we note that expressions\(^8\) and\(^9\) imply that for small incident fields, the same expression

\[
\tilde{\alpha}(\omega_L) = \frac{1}{2} \frac{\omega_{ca}}{\omega_L - \omega_{ca} + i\frac{\Gamma}{2}},
\]

(34)

for the linear dynamic polarizability\(^10\) is obtained, as it should be.

V. SUMMARY

In this paper, we have solved the optical Bloch equations for a two-level system interacting with a statistically stationary broadband field. The resulting steady-state density matrix is similar to the result found for a monochromatic incident field; the difference between both results can be intuitively understood. Finally, we have applied the obtained results to calculate the full response of a two-level atom to a broadband field, expressed by the atom’s dynamic polarizability.

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