

Appendix A

Semianalytical Derivation of the Atomic Polarizability

The purpose of this section is to derive the linear polarizability of a two-level quantum system in the dipole approximation. The quantum system might be an atom, a molecule, or a quantum dot. For simplicity, we will denote the system as atom. Once the atomic polarizability is known, the interaction between atom and radiation field can be treated in many applications classically. A generally valid analytical expression for the polarizability cannot be derived. Instead, one has to distinguish between several approximate expressions which depend on the relative spectral properties of atom and field. The two most important regimes are *off-resonance* and *near-resonance* excitation. In the former case, the atom resides mostly in its ground state whereas in the latter case, saturation of the excited level becomes significant.

According to quantum mechanics, the behavior of a system of N particles is described by the wavefunction

$$\Psi(\mathbf{r}, t) = \Psi(\mathbf{r}_1, \dots, \mathbf{r}_N, t), \quad (\text{A.1})$$

where \mathbf{r}_i denotes the spatial coordinate of particle i and t represents the time variable. To make the notation simpler, the entire set of particle coordinates is represented by the single coordinate \mathbf{r} which also includes spin. However, it should be kept in mind that operations on \mathbf{r} are operations on all particle coordinates $\mathbf{r}_1, \dots, \mathbf{r}_N$. The wavefunction Ψ is a solution of the Schrödinger equation

$$\hat{H} \Psi(\mathbf{r}, t) = i\hbar \frac{d}{dt} \Psi(\mathbf{r}, t). \quad (\text{A.2})$$

\hat{H} denotes the Hamilton operator, also called Hamiltonian. Its form depends on the considered system.

For an isolated atom with no external perturbation the Hamiltonian is time independent and it has the general form

$$\hat{H}_o = \sum_{i,j} \left[-\frac{\hbar^2}{2m_i} \nabla_i^2 + V(\mathbf{r}_i, \mathbf{r}_j) \right]. \quad (\text{A.3})$$

The sum runs over all particles involved in the system. The index in ∇_i specifies operation on the coordinate \mathbf{r}_i . $V(\mathbf{r}_i, \mathbf{r}_j)$ is the potential interaction energy of the i th and j th particle. In general, V has contributions from all four fundamental interactions so far known, namely strong, electromagnetic, weak, and gravitational interaction. For the behaviour of electrons only the electromagnetic contribution is of importance, and within the electromagnetic interaction the electrostatic potential is of dominance. Since the masses of nuclei are much greater than the mass of an electron, the nuclei move much slower than the electrons. This allows the electrons to practically instantaneously follow the nuclear motion. For an electron, the nucleus appears to be at rest. This is the essence of the Born-Oppenheimer approximation which allows us to separate the nuclear wavefunction from the electronic one. We therefore consider a nucleus of total charge qZ , Z being the atomic number. We assume that the nucleus is located at the origin of coordinates ($\mathbf{r} = 0$) surrounded by Z electrons each of charge $-q$. We can restrict the index i in Eq. (A.3) to run only over electron coordinates. In the case of a time independent Hamiltonian we can separate the t and \mathbf{r} dependence as

$$\Psi(\mathbf{r}, t) = \sum_{n=1}^{\infty} e^{-i/\hbar E_n t} \varphi_n(\mathbf{r}). \quad (\text{A.4})$$

Inserting this wavefunction into Eq. (A.2) and using $\hat{H} = \hat{H}_o$ we obtain the energy eigenvalue equation (time-independent Schrödinger equation)

$$\hat{H}_o \varphi_n(\mathbf{r}) = E_n \varphi_n(\mathbf{r}), \quad (\text{A.5})$$

where E_n are the energy eigenvalues of the stationary states $|n\rangle$. In the following we restrict ourselves to the case of a two-level atom ($n = [1, 2]$) with the two stationary wavefunctions

$$\begin{aligned} \Psi_1(\mathbf{r}, t) &= e^{-i/\hbar E_1 t} \varphi_1(\mathbf{r}) \\ \Psi_2(\mathbf{r}, t) &= e^{-i/\hbar E_2 t} \varphi_2(\mathbf{r}). \end{aligned} \quad (\text{A.6})$$

In a next step, we expose the atomic system to the radiation field. The system then experiences an external, time dependent perturbation represented by the interaction Hamiltonian $\hat{H}'(t)$. We obtain for the total Hamiltonian

$$\hat{H} = \hat{H}_o + \hat{H}'(t), \quad (\text{A.7})$$

where \hat{H}_o represents the unperturbed system according to Eq. (A.5). The size of an atom is on the order of a couple of Bohr radii, $a_B \approx 0.05 \text{ nm}$. Since $a_B \ll \lambda$, λ being the

wavelength of the radiation field, we can assume that the electric field \mathbf{E} is constant across the dimensions of the atomic system. Assuming time-harmonic fields, we can write

$$\mathbf{E}(\mathbf{r}, t) = \text{Re} \{ \mathbf{E}(\mathbf{r}) e^{-i\omega t} \} \approx \mathbf{E}_o \cos(\omega t), \quad (\text{A.8})$$

where we have set the phase of the field equal to zero or, equivalently, we have chosen the complex field amplitude to be real. Each electron in the system experiences the same field strength \mathbf{E}_o and the same time dependence $\cos(\omega t)$. Using the total electric dipole moment of the atom

$$\boldsymbol{\mu}_a(\mathbf{r}) = \boldsymbol{\mu}_a(\mathbf{r}_1, \dots, \mathbf{r}_Z) = q \sum_{i=1}^Z \mathbf{r}_i \quad (\text{A.9})$$

we find for the interaction Hamiltonian in the dipole approximation

$$\hat{H}' = -\boldsymbol{\mu}_a(\mathbf{r}) \cdot \mathbf{E}_o \cos(\omega t). \quad (\text{A.10})$$

The dipolar interaction Hamiltonian is real and has odd parity, i.e. \hat{H}' changes sign if the inversion operation $\mathbf{r}_i = -\mathbf{r}_i$ is applied to all \mathbf{r}_i .

To solve the Schrödinger equation (A.2) for the perturbed system we make a time dependent superposition of the stationary atomic wavefunctions in Eq. (A.6) as

$$\Psi(\mathbf{r}, t) = c_1(t)\Psi_1(\mathbf{r}, t) + c_2(t)\Psi_2(\mathbf{r}, t). \quad (\text{A.11})$$

We choose the time dependent coefficients c_1 and c_2 such that the normalization condition $\langle \Psi | \Psi \rangle = \int \Psi^* \Psi dV = |c_1|^2 + |c_2|^2 = 1$ is fulfilled. For clarity, we will drop the arguments in the wavefunctions. After inserting this wavefunction into Eq. (A.2), rearranging terms and making use of Eq. (A.3) and Eqs. (A.6) we obtain

$$\hat{H}'(c_1\Psi_1 + c_2\Psi_2) = i\hbar [\Psi_1 \dot{c}_1 + \Psi_2 \dot{c}_2], \quad (\text{A.12})$$

where the dots denote differentiation with respect to time. It should be kept in mind, that the arguments of Ψ and φ are (\mathbf{r}, t) and (\mathbf{r}) , respectively. To eliminate the spatial dependence we multiply Eq. (A.12) from the left by Ψ_1^* on both sides, introduce expressions A.6 for the wavefunctions and integrate over all space. After repeating the procedure with Ψ_2^* instead of Ψ_1^* we obtain a set of two time-dependent coupled differential equations

$$\dot{c}_1(t) = c_2(t) \frac{i}{\hbar} \boldsymbol{\mu}_{12} \cdot \mathbf{E}_o \cos(\omega t) e^{-i/\hbar(E_2-E_1)t} \quad (\text{A.13})$$

$$\dot{c}_2(t) = c_1(t) \frac{i}{\hbar} \boldsymbol{\mu}_{21} \cdot \mathbf{E}_o \cos(\omega t) e^{+i/\hbar(E_2-E_1)t}. \quad (\text{A.14})$$

We have introduced the definition of the dipole matrix element between the states $|i\rangle$ and $|j\rangle$ as

$$\boldsymbol{\mu}_{ij} = \langle i | \boldsymbol{\mu}_a | j \rangle = \int \varphi_i^*(\mathbf{r}) \boldsymbol{\mu}_a(\mathbf{r}) \varphi_j(\mathbf{r}) dV, \quad (\text{A.15})$$

It has to be emphasized again that the integration runs over all electron coordinates $\mathbf{r} = \mathbf{r}_1, \dots, \mathbf{r}_Z$. In Eqs. (A.13) and (A.14) we have used the fact that $\mathbf{p}_{ii} = 0$. This follows from the odd parity of \hat{H}' which makes the integrands of $\boldsymbol{\mu}_{ii}$ odd functions of \mathbf{r} . Integration over $\mathbf{r} = [-\infty \dots 0]$ leads to the opposite result than integration over $\mathbf{r} = [0 \dots \infty]$. Upon integration over all space the two contributions cancel. The dipole matrix elements satisfy $\boldsymbol{\mu}_{12} = \boldsymbol{\mu}_{21}^*$ because $\boldsymbol{\mu}_a$ is a Hermitian operator. However, it is convenient to choose the phases of the eigenfunctions φ_1 and φ_2 such that the dipole matrix elements are real, i.e.

$$\boldsymbol{\mu}_{12} = \boldsymbol{\mu}_{21} . \quad (\text{A.16})$$

In the following, we will assume that $\Delta E = E_2 - E_1 > 0$, and we introduce the *transition frequency*

$$\omega_o = \Delta E / \hbar . \quad (\text{A.17})$$

for the sake of simpler notation. The state $|1\rangle$ is the ground state and the state $|2\rangle$ the excited state.

Semiclassical theory does not account for spontaneous emission. The spontaneous emission process can only be found by use of a quantized radiation field whose Hamiltonian has to be included in Eq. (A.7). To be in accordance with quantum electrodynamics we have to include the effects of spontaneous emission by introducing a phenomenological damping term in Eq. (A.14). The coupled differential equations have then the form

$$\begin{cases} \dot{c}_1(t) = c_2(t) \, i/\hbar \, \boldsymbol{\mu}_{12} \cdot \mathbf{E}_o \cos(\omega t) e^{-i\omega_o t} \\ \dot{c}_2(t) + \gamma/2 c_2(t) = c_1(t) \, i/\hbar \, \boldsymbol{\mu}_{21} \cdot \mathbf{E}_o \cos(\omega t) e^{+i\omega_o t} \end{cases} . \quad (\text{A.18})$$

The introduction of the damping term asserts that an excited atom must ultimately decay to its ground state by spontaneous emission. In the absence of the radiation field, $\mathbf{E}_o = 0$, Eq. (A.18) can be integrated at once and we obtain

$$c_2(t) = c_2(0) e^{-\gamma/2 t} . \quad (\text{A.19})$$

The average lifetime τ of the excited state is $\tau = 1/\gamma$, γ being the spontaneous decay rate. Since there is no direct analytical solution of Eqs. (A.18) we have to find approximate solutions for different types of excitations.

Steady state polarizability for weak excitation fields

We assume that the interaction between atom and radiation field is weak. The solution for $c_1(t)$ and $c_2(t)$ can then be represented as a power series in $\boldsymbol{\mu}_{21} \cdot \mathbf{E}_o$. To derive the first order term in this series we set $c_1(t) = 1$ and $c_2(t) = 0$ on the right hand side of

Eqs. (A.18). Once we have found the first order solution we can insert it again to the right hand side to find the second order solution and so on. However, we will restrict ourselves to the first order term. The solution for c_1 is $c_1(t) = 1$ indicating that the atom resides always in its ground state. This solution is the zero order solution, i.e. there is no first order solution for c_1 . The next higher term would be of second order. The first order solution for c_2 is obtained by a superposition of the homogeneous solution in Eq. (A.19) and a particular solution. The latter is easily found by writing the cosine term as a sum of two exponentials. We then obtain for the first order solution of c_2

$$c_2(t) = \boldsymbol{\mu}_{21} \cdot \mathbf{E}_o \frac{1}{2\hbar} \left[\frac{e^{i(\omega_o + \omega)t}}{\omega_o + \omega - i\gamma/2} + \frac{e^{i(\omega_o - \omega)t}}{\omega_o - \omega - i\gamma/2} \right] + c_2(0) e^{-\gamma/2 t}. \quad (\text{A.20})$$

We are interested in calculating the *steady state* behavior for which the atom has been subjected to the electric field $\mathbf{E}_o \cos(\omega t)$ for an infinitely long period of time. In this situation the inhomogeneous term disappears and the solution is given by the homogeneous solution alone.

The expectation value of the dipole moment is defined as

$$\boldsymbol{\mu}(t) = \langle \Psi | \boldsymbol{\mu}_a | \Psi \rangle = \int \Psi^*(\mathbf{r}) \boldsymbol{\mu}_a(\mathbf{r}) \Psi(\mathbf{r}) dV, \quad (\text{A.21})$$

The integration runs again over all coordinates \mathbf{r}_i . Using the wavefunction Ψ of Eq. (A.11) the expression for $\boldsymbol{\mu}$ becomes

$$\boldsymbol{\mu}(t) = c_1^* c_2 \boldsymbol{\mu}_{12} e^{-i\omega_o t} + c_1 c_2^* \boldsymbol{\mu}_{21} e^{i\omega_o t}, \quad (\text{A.22})$$

where we used the definition of the dipole matrix elements of Eq. (A.15) and the property $\boldsymbol{\mu}_{ii} = 0$. Using the first order solutions for c_1 and c_2 we obtain

$$\boldsymbol{\mu}(t) = \frac{\boldsymbol{\mu}_{12} [\boldsymbol{\mu}_{21} \cdot \mathbf{E}_o]}{2\hbar} \left[\frac{e^{i\omega t}}{\omega_o + \omega - i\gamma/2} + \frac{e^{-i\omega t}}{\omega_o - \omega - i\gamma/2} + \frac{e^{-i\omega t}}{\omega_o + \omega + i\gamma/2} + \frac{e^{i\omega t}}{\omega_o - \omega + i\gamma/2} \right]. \quad (\text{A.23})$$

Since the exciting electric field is given as $\mathbf{E} = (1/2)\mathbf{E}_o[\exp(i\omega t) + \exp(-i\omega t)]$ we rewrite the dipole moment above as

$$\boldsymbol{\mu}(t) = \frac{1}{2} \left[\overleftrightarrow{\alpha}^*(\omega) e^{i\omega t} + \overleftrightarrow{\alpha}(\omega) e^{-i\omega t} \right] \mathbf{E}_o = \text{Re} \left\{ \overleftrightarrow{\alpha}(\omega) e^{-i\omega t} \right\} \mathbf{E}_o, \quad (\text{A.24})$$

where $\overleftrightarrow{\alpha}$ is the *atomic polarizability* tensor

$$\overleftrightarrow{\alpha}(\omega) = \frac{\boldsymbol{\mu}_{12} \boldsymbol{\mu}_{21}}{\hbar} \left[\frac{1}{\omega_o - \omega - i\gamma/2} + \frac{1}{\omega_o + \omega + i\gamma/2} \right]. \quad (\text{A.25})$$

$\boldsymbol{\mu}_{12} \boldsymbol{\mu}_{21}$ denotes the matrix formed by the outer product between the (real) transition dipole moments. It is convenient to write the polarizability in terms of a single

denominator. Furthermore, we realize that the damping term γ is much smaller than ω_o which allows us to drop terms in γ^2 . Finally, we have to generalize the result to a system with more than two states. Besides the different matrix elements, each state different from the ground state behaves in a similar way than our previous state $|2\rangle$. Thus, each new level is characterized by its natural frequency ω_n , its damping term γ_n and the transition dipole moments $\boldsymbol{\mu}_{1n}, \boldsymbol{\mu}_{n1}$. Then, the polarizability takes on the form

$$\boxed{\vec{\alpha}(\omega) = \sum_n \vec{\mathbf{f}}_n \left[\frac{e^2/m}{\omega_n^2 - \omega^2 - i\omega\gamma_n} \right], \quad \vec{\mathbf{f}}_n = \frac{2m\omega_n}{e^2\hbar} \boldsymbol{\mu}_{1n} \boldsymbol{\mu}_{n1}}, \quad (\text{A.26})$$

where $\vec{\mathbf{f}}_n$ is the so-called *oscillator strength** and e and m denote the electron charge and mass, respectively. It is for historical reasons that we have cast the polarizability into the form of Eq. (A.26). Before the advent of quantum mechanics, H. A. Lorentz developed a classical model for the atomic polarizability which, besides the expression for $\vec{\mathbf{f}}_n$, is identical with our result. The model considered by Lorentz consists of a collection of harmonic oscillators for the electrons of an atom. Each electron responds to the driving incident field according to the equation of motion

$$\ddot{\boldsymbol{\mu}} + \gamma\dot{\boldsymbol{\mu}} + \omega_o^2\boldsymbol{\mu} = (q^2/m) \vec{\mathbf{f}} \mathbf{E}(t). \quad (\text{A.27})$$

In this theory, the oscillator strength is a fitting parameter since there is no direct way to know how much an electron contributes to a particular atomic mode. On the other hand, the semiclassical theory directly relates the oscillator strength to the transition dipole matrix elements and thus to the atomic wavefunctions. Furthermore, the f -sum rule tells us that the sum of all oscillator strengths is equal to one.

If the energy $\hbar\omega$ of the exciting field is close to the energy difference ΔE between two atomic states, the first term in Eq. (A.25) is much larger than the second one. In this case we can discard the second term (rotating wave approximation) and the imaginary part of the polarizability becomes a perfect Lorentzian function.

It is important to notice that there is a linear relationship between the exciting electric field \mathbf{E} and the induced dipole moment $\boldsymbol{\mu}$. Therefore, a monochromatic field with angular frequency ω produces a harmonically oscillating dipole with the same frequency. This allows us to use the complex notation for $\boldsymbol{\mu}$ and \mathbf{E} and write

$$\boldsymbol{\mu} = \vec{\alpha} \mathbf{E}, \quad (\text{A.28})$$

from which we obtain the time dependence of \mathbf{E} and $\boldsymbol{\mu}$ by simply multiplying by $\exp(-i\omega t)$ and taking the real part.

*The average over all polarizations reduces the oscillator strength to a scalar quantity with an extra factor of 1/3.

Near-resonance excitation in absence of damping

In the previous section we required that the interaction between excitation beam and the atom is weak and that the atom resides mostly in its ground state. This condition can be relaxed if we consider an exciting field whose energy $\hbar\omega$ is close to the energy difference ΔE between two atomic states. As mentioned before, there is no direct analytical solution to the coupled differential equations in Eqs. (A.18). However, a quite accurate solution can be found if we drop the damping term γ and if the energy of the radiation field is close to the energy difference between excited and ground state, i.e.

$$|\hbar\omega - \Delta E| \ll \hbar\omega + \Delta E . \quad (\text{A.29})$$

In this case, we can apply the so-called rotating wave approximation. After rewriting the cosines in Eqs. (A.18) in terms of exponentials we find exponents with $(\hbar\omega \pm \Delta E)$. In the rotating wave approximation we only retain terms with $(\hbar\omega - \Delta E)$ because of their dominating contributions. Eqs. (A.18) then become[†]

$$\frac{i}{2} \omega_R e^{-i(\omega_o - \omega)t} c_2(t) = \dot{c}_1(t) \quad (\text{A.30})$$

$$\frac{i}{2} \omega_R e^{i(\omega_o - \omega)t} c_1(t) = \dot{c}_2(t) , \quad (\text{A.31})$$

where we introduced the *Rabi frequency* ω_R defined as

$$\omega_R = \frac{|\boldsymbol{\mu}_{12} \cdot \mathbf{E}_o|}{\hbar} = \frac{|\boldsymbol{\mu}_{21} \cdot \mathbf{E}_o|}{\hbar} . \quad (\text{A.32})$$

ω_R is a measure for the strength of the time-varying external field. Inserting the trial solution $c_1(t) = \exp(i\kappa t)$ into the first equation (A.30) we find $c_2(t) = (2\kappa/\omega_R) \exp(i[\omega_o - \omega + \kappa]t)$. Substituting both c_1 and c_2 into the second equation (A.31) we find a quadratic equation for the unknown parameter κ leading to the two solutions κ_1 and κ_2 . The general solutions for the amplitudes c_1 and c_2 can then be written as

$$c_1(t) = A e^{i\kappa_1 t} + B e^{i\kappa_2 t} \quad (\text{A.33})$$

$$c_2(t) = (2/\omega_R) e^{i(\omega_o - \omega)t} [A\kappa_1 e^{i\kappa_1 t} + B\kappa_2 e^{i\kappa_2 t}] . \quad (\text{A.34})$$

To determine the constants A and B we have to require appropriate boundary conditions. The probability for finding the atomic system in the excited state $|2\rangle$ is $|c_2|^2$. Similarly, the probability for finding the atom in its ground state $|1\rangle$ is $|c_1|^2$. By using the boundary conditions for the atom being initially in its ground state

$$\begin{aligned} |c_1(t=0)|^2 &= 1 \\ |c_2(t=0)|^2 &= 0 , \end{aligned} \quad (\text{A.35})$$

[†]We again choose the phases of the atomic wavefunctions such that the transition dipole matrix elements are real.

the unknown constants A and B can be determined. Using the expressions for κ_1 , κ_2 , A , and B , we finally find the solution

$$c_1(t) = e^{-i/2(\omega_o-\omega)t} \left[\cos(\Omega t/2) - \frac{i(\omega-\omega_o)}{\Omega} \sin(\Omega t/2) \right] \quad (\text{A.36})$$

$$c_2(t) = \frac{i\omega_R}{\Omega} e^{i/2(\omega_o-\omega)t} \sin(\Omega t/2) , \quad (\text{A.37})$$

where Ω denotes the *Rabi-flopping frequency* defined as

$$\Omega = \sqrt{(\omega_o-\omega)^2 + \omega_R^2} . \quad (\text{A.38})$$

It can be easily shown that $|c_1|^2 + |c_2|^2 = 1$. The probability for finding the atom in its excited state becomes

$$|c_2(t)|^2 = \omega_R^2 \frac{\sin^2(\Omega t/2)}{\Omega^2} . \quad (\text{A.39})$$

The transition probability is a periodic function of time. The system oscillates between the levels E_1 and E_2 at the frequency $\Omega/2$ which depends on the detuning $\omega_o-\omega$ and the field strength represented by ω_R . If ω_R is small we have $\Omega \approx (\omega_o-\omega)$ and, in the absence of damping, the results become identical with the results of the previous section.

The expectation value of the dipole moment is defined by Eqs. (A.21) and Eq. (A.22). Inserting the solutions for c_1 and c_2 and using Eq. (A.16) we obtain

$$\boldsymbol{\mu}(t) = \boldsymbol{\mu}_{12} \frac{\omega_R}{\Omega} \left[\frac{(\omega-\omega_o)}{\Omega} [1-\cos(\Omega t)] \cos(\omega t) + \sin(\Omega t) \sin(\omega t) \right] . \quad (\text{A.40})$$

We see that the induced dipole moment oscillates at the frequency of the radiation field. However, it does not instantaneously follow the driving field: it has in-phase and quadrature components. Let us write $\boldsymbol{\mu}$ in the complex representation as

$$\boldsymbol{\mu}(t) = \text{Re} \{ \boldsymbol{\mu} e^{-i\omega t} \} . \quad (\text{A.41})$$

We then find for the complex dipole moment

$$\boldsymbol{\mu} = \boldsymbol{\mu}_{12} \frac{\omega_R}{\Omega} \left[\frac{(\omega-\omega_o)}{\Omega} [1-\cos(\Omega t)] + i \sin(\Omega t) \right] . \quad (\text{A.42})$$

To determine the atomic polarizability, defined as

$$\boldsymbol{\mu} = \overset{\leftrightarrow}{\alpha} \mathbf{E} , \quad (\text{A.43})$$

we have to express the Rabi frequency ω_R by its definition Eq. (A.32) and obtain

$$\boxed{\overset{\leftrightarrow}{\alpha}(\omega) = \frac{\boldsymbol{\mu}_{12} \boldsymbol{\mu}_{21}}{\hbar} \left[\frac{(\omega-\omega_o)}{\Omega^2} [1-\cos(\Omega t)] + i \sin(\Omega t) \right]} . \quad (\text{A.44})$$

The most remarkable property of the polarizability is its dependence on field strength (through ω_R) and its time-dependence. This is different to the polarizability derived in the previous section. In the present case, the time-behavior is determined by the Rabi flopping frequency Ω . In practical situations the time-dependence disappears within tens of nanoseconds because of the damping term γ which has been neglected in our derivation. For the case of exact resonance ($\omega = \omega_o$) the polarizability reduces to a sinusoidal function of $\omega_R t$. This oscillation is much slower than the oscillation of the optical resonance. For weak interactions ω_R is small and the polarizability becomes a linear function of t .

Near-resonance excitation with damping

The damping term γ attenuates the purely oscillatory solution derived in the previous section. After a sufficiently long time, the system will relax into the ground-state. To calculate the steady state behavior it is sufficient to solve for the term $c_1 c_2^*$ which, together with its complex conjugate, defines the expectation value of the dipole moment [c.f. Eq. (A.22)]. In the steady-state, the probability of finding the atom in its excited state will be time-independent, i.e.

$$\frac{d}{dt} [c_2 c_2^*] = 0 \quad (\text{steady - state}) . \quad (\text{A.45})$$

Furthermore, in the rotating-wave approximation, it can be expected that the time-dependence of the off-diagonal matrix element $c_1 c_2^*$ will be solely defined by the factor $\exp(-i[\omega_o - \omega]t)$. Thus,

$$\frac{d}{dt} [c_1 c_2^*] = -i(\omega_o - \omega) [c_1 c_2^*] \quad (\text{steady - state}) , \quad (\text{A.46})$$

with a similar equation for $c_2 c_1^*$. Using

$$\frac{d}{dt} [c_i c_j^*] = c_i \dot{c}_j^* + c_j^* \dot{c}_i , \quad (\text{A.47})$$

inserting Eqs. (A.18), applying the rotating-wave approximation, and making use of the steady-state conditions above, we obtain

$$\omega_R \exp(-i[\omega_o - \omega]t) [c_2 c_1^*] - \omega_R^* \exp(i[\omega_o - \omega]t) [c_1 c_2^*] - 2i\gamma [c_2 c_2^*] = 0 \quad (\text{A.48})$$

$$\omega_R ([c_1 c_1^*] - [c_2 c_2^*]) - (2[\omega_o - \omega] + i\gamma) \exp(i[\omega_o - \omega]t) [c_1 c_2^*] = 0 \quad (\text{A.49})$$

$$\omega_R ([c_1 c_1^*] - [c_2 c_2^*]) - (2[\omega_o - \omega] - i\gamma) \exp(i[\omega_o - \omega]t) [c_2 c_1^*] = 0 \quad (\text{A.50})$$

This set of equations can be solved for $[c_1 c_2^*]$ and gives

$$[c_1 c_2^*] = e^{-i(\omega_o - \omega)t} \frac{1/2 \omega_R (\omega_o - \omega - i\gamma/2)}{(\omega_o - \omega)^2 + \gamma^2/4 + 1/2 \omega_R^2} , \quad (\text{A.51})$$

with the complex conjugate solution for $[c_2 c_1^*]$. The expectation value of the dipole moment can now be calculated by using Eq. (A.22) and the steady-state solution for the atomic polarizability for near-resonance excitation ($\omega \approx \omega_o$) can be determined as

$$\boxed{\vec{\alpha}(\omega) = \frac{\boldsymbol{\mu}_{12} \boldsymbol{\mu}_{21}}{\hbar} \frac{(\omega_o - \omega + i\gamma/2)}{(\omega_o - \omega)^2 + \gamma^2/4 + 1/2\omega_R^2}}. \quad (\text{A.52})$$

The most remarkable difference to the off-resonant case is the appearance of the term ω_R^2 in the denominator. This term accounts for saturation of the excited state thereby reducing the absorption rate and increasing the linewidth from γ to $(\gamma + 2\omega_R^2)^{1/2}$ which is denoted as *saturation broadening*. Thus, the damping constant becomes dependent on the acting electric field strength. Saturation is not a nonlinear behavior in the usual sense since the dipole moment $\boldsymbol{\mu}$ has always the same harmonic time-dependence as the driving electric field. Saturation in the steady-state gives only rise to a nonlinear relationship between the amplitudes of dipole moment and electric field. For $\omega_R \rightarrow 0$, the polarizability reduces to

$$\vec{\alpha}(\omega) = \frac{\boldsymbol{\mu}_{12} \boldsymbol{\mu}_{21}}{\hbar} \frac{1}{\omega_o - \omega - i\gamma/2}, \quad (\text{A.53})$$

which is identical with the rotating-wave term of Eq. (A.25).

The polarizability can be calculated once the energy levels E_1 and E_2 and the dipole matrix element $\boldsymbol{\mu}_{12}$ are known. The latter is defined by Eq. (A.15) through the wavefunctions φ_1 and φ_2 . It is thus necessary to solve the energy eigenvalue equation (A.5) for the considered quantum system in order to accurately determine the energy levels and the dipole matrix element. However, Eq. (A.5) can be solved analytically only for simple systems often restricted to two interacting particles. Systems with more than two interacting particles have to be treated with approximate methods such as the Hartree-Fock method or numerically.