# Chapter 2
# Theoretical Model

In this chapter, the theoretical models used in the thesis are briefly introduced. The system composed of a two-level quantum resonator, e.g., nucleus or atom interacting with a single mode of the electromagnetic wave is a basic but nontrivial problem. Many important properties of matter-field interaction can be extracted from it. For this reason we start with an example involving the coupling of a closed two-level nucleus interacting with a single-mode radiation field. We first present the used equations of motion for density matrices in Sect. 2.1. The main purpose of Sect. 2.1 is to derive the form of the Hamiltonian matrix elements adopted in nuclear physics. Furthermore, by coupling the equations for the density matrix to the Maxwell equations (i.e, Maxwell-Bloch equation) as shown in Sect. 2.2, one can study the fruitful physics of the propagation of a light pulse through a resonant medium. This issue focuses on solving the dynamics of the incident electromagnetic wave which is associated with probing a material sample from the the photons scattered off a target, e.g., as in the nuclear forward scattering setup [1]. We present the used theory in Sect. 2.2 with a very useful example about the interaction of a short light pulse with two-level nuclei. In Sect. 2.2.1, an analytic solution of the dynamics of the incident pulse is derived via Maxwell-Bloch equations confirming an expression frequently appearing in the corresponding literature as a result of a different theoretical approach. Finally, in Sect. 2.3 we demonstrate an extension of the used theory towards a three-level $\Lambda$-type system [2] which is well known for some topics in atomic quantum optics, e.g., stimulated Raman adiabatic passage (STIRAP) [3] and electromagnetically induced transparency (EIT) [4–6].

## 2.1 Master Equation

In this thesis, the master equation[1] is used to describe the dynamics of the considered nuclei-radiation system:

[1] Also called optical Bloch equation or Liouville-von Neumann equation in literature [2].

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$$\partial_t \widehat{\rho} = \frac{1}{i\hbar}\left[\widehat{H}, \widehat{\rho}\right] + \widehat{\rho}_s. \tag{2.1}$$

Equation (2.1) describes the quantum time evolution of the density operator $\widehat{\rho}(t)$ of matter in a system, e.g., nuclei in this thesis. $\widehat{H}(t)$ is the interaction Hamiltonian between the matter and the external fields, e.g., incident electromagnetic fields, and $\widehat{\rho}_s$ describes the decoherence processes such as spontaneous decay. Moreover, Eq. (2.1) is very general for any quantum system, and the way to calculate the matrix elements of $\widehat{H}(t)$ depends on the considered physical problems. In the following, we present an example for a two-level system, and the form of $\widehat{H}(t)$ matrix elements used in nuclear physics.

A typical two-level system is illustrated in Fig. 2.1. The interaction between a nucleus and a pump laser is illustrated in Fig. 2.1c Here, pump laser (blue arrow) drives the transition $|2\rangle \leftrightarrow |1\rangle$, with detuning $\Delta_p$. The explicit form of $\widehat{\rho}$ is:

$$\widehat{\rho} = \begin{pmatrix} \rho_{11} & \rho_{12} \\ \rho_{21} & \rho_{22} \end{pmatrix}, \tag{2.2}$$

for a two-level nuclear wavefunction $|\psi\rangle = C_1(t)|1\rangle + C_2(t)|2\rangle$

$$\rho_{eg} = C_e C_g^*. \tag{2.3}$$

Here indices $e, g \in \{1, 2\}$. Considering the spontaneous decay for the nuclear system, the decoherence matrix is

$$\widehat{\rho}_s = \frac{\Gamma}{2}\begin{pmatrix} 2\rho_{22} & -\rho_{12} \\ -\rho_{21} & -2\rho_{22} \end{pmatrix}. \tag{2.4}$$

In Eq. (2.4), the relation $\rho_{11} + \rho_{22} = 1$ is satisfied due to the conservation of population in a closed two-level system. The decay rate $\Gamma/2$ for each off diagonal coherence is derived from Eq. (2.3). Without any external laser, $\rho_{22}(t) \sim e^{-\Gamma t}$ which means $C_2(t) \sim e^{-(\Gamma/2)t}$, whence the other coherence $\rho_{21}$ and $\rho_{12}$ are also proportional to $e^{-(\Gamma/2)t}$. Furthermore, in the interaction picture, the interaction Hamiltonian matrix $\widehat{H}(t)$ is [3]:

$$\widehat{H} = -\frac{\hbar}{2}\begin{pmatrix} 0 & \Omega_p^* \\ \Omega_p & 2\Delta_p \end{pmatrix}, \tag{2.5}$$

where $\hbar$ is the reduced Planck constant, and $\Delta_p$ is the laser detuning. The explicit form of Eq. (2.1) can be obtained by substituting Eqs. (2.3), (2.4) and (2.5) into Eq. (2.1):

$$\partial_t \rho_{11} = \Gamma \rho_{22} + \frac{i}{2}\left(\Omega_p^* \rho_{21} - \Omega_p \rho_{21}^*\right) \tag{2.6}$$

$$\partial_t \rho_{21} = -\left(\frac{\Gamma}{2} + i\Delta_p\right)\rho_{21} - \frac{i}{2}\Omega_p\left(\rho_{22} - \rho_{11}\right) \tag{2.7}$$

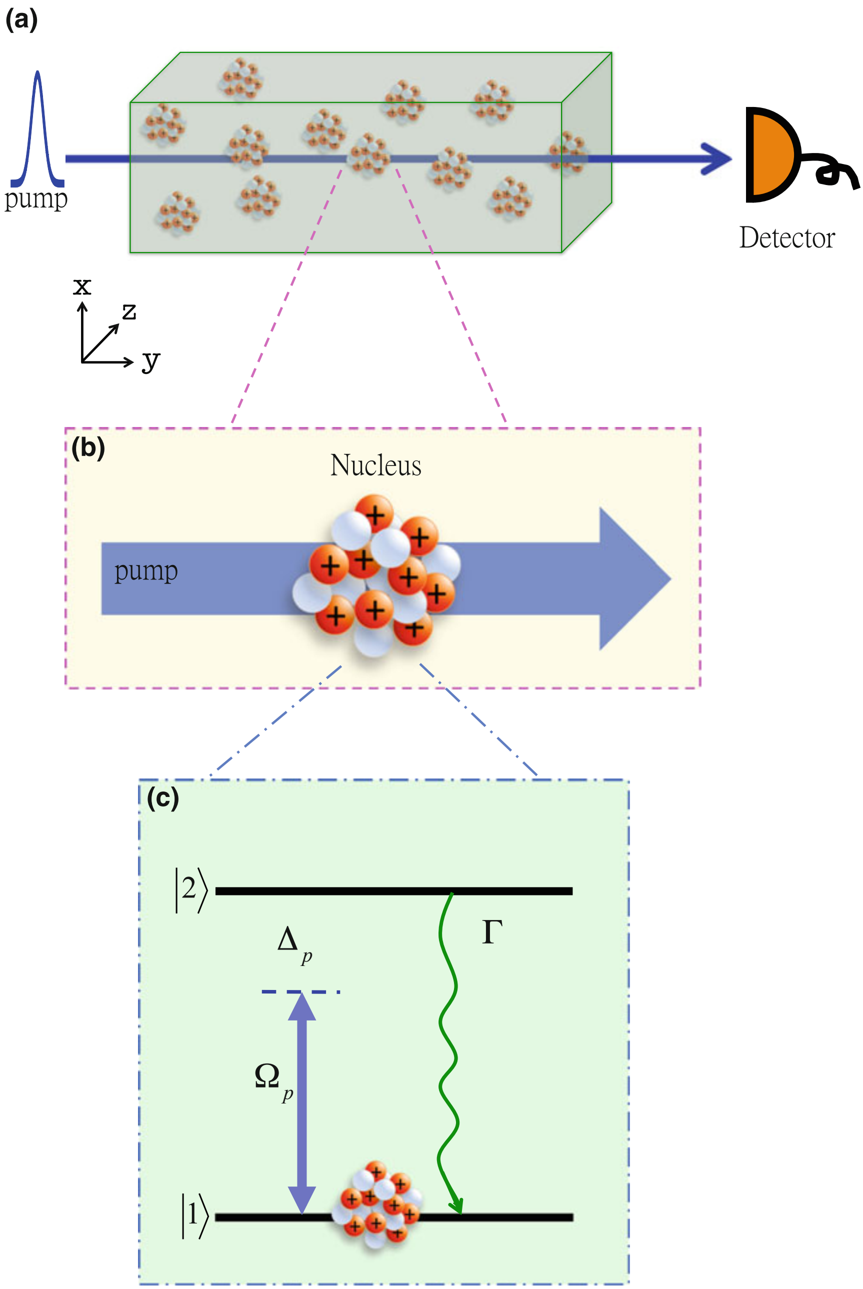


**Fig. 2.1** **a** Illustration of coherent pulse propagation through a resonant medium. **b** Interaction between a nucleus and pump laser (*blue wide arrow*). **c** Sketch of a two-level nuclear system. The *blue* ($\Omega_p$) *arrow* depicts the pump laser, and $\Delta_p$ denotes the pump laser detuning. The *green wiggled arrow* illustrates the spontaneous decay of state $|2\rangle$ with a rate of $\Gamma$

$$\partial_t \rho_{22} = -\Gamma \rho_{22} + \frac{i}{2}\left(\Omega_p \rho_{21}^* - \Omega_p^* \rho_{21}\right). \tag{2.8}$$

In the equations above, $\Omega_p$ denotes Rabi frequency defined as:

$$\Omega_p(t) = \frac{1}{\hbar}\langle 2|\widehat{H}_I(t)|1\rangle, \tag{2.9}$$

and by using the Coulomb gauge for the vector potential of pump laser $A = \sum_k A_k e^{i(kr-\omega_k t)}$ + H.c., the matrix element can be written as

$$\langle e|\widehat{H}_I(t)|g\rangle = -\langle e|\widehat{\mathbf{j}}\cdot\mathbf{A}(t)|g\rangle \tag{2.10}$$

$$= -\sum_k \langle e|\left[\widehat{\mathbf{j}}e^{-i\omega_{eg}t} + \widehat{\mathbf{j}}^* e^{i\omega_{eg}t}\right] \cdot \left[\mathbf{A_k} e^{i(\mathbf{kr}-\omega_k t)} + \mathbf{A_k^*} e^{-i(\mathbf{kr}-\omega_k t)}\right]|g\rangle \tag{2.11}$$

where $\widehat{\mathbf{j}}$ is nuclear current operator. By using the rotating wave approximation [2] and considering the interaction between nuclei and a single $k$ mode plane wave, Eq. (2.11) becomes

$$\langle e|\widehat{H}_I(t)|g\rangle = -\langle e|\left\{\widehat{\mathbf{j}}\cdot\mathbf{A_k^*}e^{-i[\mathbf{k}\cdot\mathbf{r}+(\Omega_{eg}-\Omega_k)t]} + \widehat{\mathbf{j}}^*\cdot\mathbf{A_k}e^{i[\mathbf{k}\cdot\mathbf{r}+(\omega_{eg}-\omega_k)t]}\right\}|g\rangle \tag{2.12}$$

$$= -\langle e|\left\{\widehat{\mathbf{j}}\cdot\mathbf{A_k^*}e^{-i(\mathbf{k}\cdot\mathbf{r}-\Delta_\mathbf{k}t)} + \widehat{\mathbf{j}}^*\cdot\mathbf{A_k}e^{i(\mathbf{k}\cdot\mathbf{r}-\Delta_\mathbf{k}t)}\right\}|g\rangle \tag{2.13}$$

$$= -e^{-i\Delta_k t}\mathbf{A_k^*}\cdot\int_V \mathbf{j}(\mathbf{r})e^{-i\mathbf{k}\cdot\mathbf{r}}d^3\mathbf{r} + H.c. \tag{2.14}$$

$$= \frac{i}{\omega_k}e^{-i\Delta_k t}\mathbf{E_k^*}\cdot\int_V \mathbf{j}(\mathbf{r})e^{-i\mathbf{k}\cdot\mathbf{r}}d^3\mathbf{r} + H.c. \tag{2.15}$$

$$= \frac{i}{\omega_k}e^{-i\Delta_k t}\mathbf{E}_k^*\cdot\int_V \mathbf{j}(\mathbf{r})e^{-ikr\cos\beta}d^3 r + H.c.. \tag{2.16}$$

Here, the laser electric field $\mathbf{E_k}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega_k t)} = -\partial_t\left[\mathbf{A_k}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega_k t)}\right] \cong i\omega_k\mathbf{A_k}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega_k t)}$ with the assumption of $|\partial_t\mathbf{A_k}| \ll |\omega_k\mathbf{A_k}|$, $\mathbf{j}(\mathbf{r})$ is the current,[2] $\Delta_k = \Delta_p$ the laser detuning and $\beta$ the angle between $\mathbf{A_k}$ and the particle position vector $\mathbf{r}$. Typically, the major task is to derive the Rabi frequency in Eq. (2.9) with some particular $\widehat{H}_I$ when using Eq. (2.1) to describe different physical problems.

In nuclear physics, due to the spherical symmetry of nuclei (just like atoms), the nuclear Hamiltonian of Eq. (2.16) is typically expressed in terms of the so called vector spherical harmonics $Y_{L\ell}^M(\theta,\phi)$ [7]. To derive a useful form for Eq. (2.16), we

[2] The current $j(r) = -i\frac{\wp\hbar}{2m}\left[\psi_\mathbf{e}^*(\mathbf{r})\nabla\psi_\mathbf{g}(\mathbf{r}) - \psi_\mathbf{g}(\mathbf{r})\nabla\psi_\mathbf{e}^*(\mathbf{r})\right]$, and the density $\rho(\mathbf{r}) = \wp\psi_\mathbf{e}^*\psi_\mathbf{g}$, where $\wp$ is the charge and $m$ is mass of the charged particle [7].

follow the steps in Ref. [7, 8] to expand the plane wave $\mathbf{E}_k e^{ik\cos\beta}$ into multipole fields. The vector spherical harmonics $\mathbf{Y}_{L\ell}^{M}(\theta,\phi)$ is introduced as following [7]:

$$\mathbf{Y}_{L\ell}^{M}(\theta,\phi) \equiv \sum_{m=-\ell}^{\ell}\sum_{n=-1}^{1} \mathrm{C}_{\ell 1}(L,M;m,n)Y_{\ell m}(\theta,\phi)\widehat{\chi}_n \tag{2.17}$$

$$\widehat{\chi}_1 = -\frac{1}{\sqrt{2}}(\widehat{e}_x + i\widehat{e}_y) \tag{2.18}$$

$$\widehat{\chi}_0 = \widehat{e}_z \tag{2.19}$$

$$\widehat{\chi}_{-1} = \frac{1}{\sqrt{2}}(\widehat{e}_x - i\widehat{e}_y), \tag{2.20}$$

where $M = m+n$, and $Y_{\ell m}$ and $\mathrm{C}_{\ell 1}(L,M;m,n)$ are the scalar spherical harmonics and the Clebsch-Gordan coefficient (as defined in [7] or see Table A.1 in Appendix A), respectively, and $\widehat{e}_\mu$ ($\widehat{\chi}_\mu$) is the unit vector in Cartesian (spherical) coordinates. For an arbitrary vector function $R(r)\mathbf{V}(\theta,\phi)$, it can be expanded into a series:

$$\mathbf{V}(\theta,\phi) = \sum_{L=0}^{\infty}\sum_{M=-L}^{L}\sum_{\ell=L-1}^{L+1} q(L,M,\ell)\mathbf{Y}_{L\ell}^{M}(\theta,\phi) \tag{2.21}$$

$$= \sum_{L=0}^{\infty}\sum_{M=-L}^{L} \left[f(L,M)\mathbf{Y}_{LL}^{M} + g(L,M)\mathbf{Y}_{LL+1}^{M} + h(L,M)\mathbf{Y}_{LL-1}^{M}\right] \tag{2.22}$$

$$f(L,M) = \int_0^{2\pi}\int_0^{\pi} \mathbf{Y}_{LL}^{M*}(\theta,\phi)\cdot\mathbf{V}(\theta,\phi)\sin\theta d\theta d\phi \tag{2.23}$$

$$g(L,M) = \int_0^{2\pi}\int_0^{\pi} \mathbf{Y}_{LL+1}^{M*}(\theta,\phi)\cdot\mathbf{V}(\theta,\phi)\sin\theta d\theta d\phi \tag{2.24}$$

$$h(L,M) = \int_0^{2\pi}\int_0^{\pi} \mathbf{Y}_{LL-1}^{M*}(\theta,\phi)\cdot\mathbf{V}(\theta,\phi)\sin\theta d\theta d\phi, \tag{2.25}$$

First, we present the expansion of a plane wave into spherical waves:

$$e^{i\mathbf{k}\cdot\mathbf{r}} = e^{ikr\cos\beta} \tag{2.26}$$

$$= \sum_{L=0}^{\infty} Y_{L0}(\beta)\int_0^{2\pi}\int_0^{\pi} Y_{L0}^{*}(\theta)\, e^{-ikr\cos\theta}\sin\theta d\theta d\phi \tag{2.27}$$

$$= \sum_{L=0}^{\infty}(i)^L j_L(kr)\sqrt{4\pi(2L+1)}Y_{L0}(\beta) \tag{2.28}$$

$$= \sum_{L=0}^{\infty} R(L;r) Y_{L0}(\beta) \tag{2.29}$$

where $j_1(z)$ is the spherical Bessel function of first kind. Second, Eq. (2.29) shows that the field to be transformed is proportional to $e^{ikr\cos\beta}\delta_{\pm 1n}\widehat{\chi}_n$, and in the following we use Eqs. (2.17–2.25) to express it in terms of vector spherical harmonics.

$$e^{ikr\cos\beta}\widehat{\chi}_n = \sum_{L'=0}^{\infty}\sum_{M=-L'}^{L'} \left[ f_n(L',M;r)\mathbf{Y}^{M}_{L'L'} + g_n(L',M;r)\mathbf{Y}^{M}_{L'L'+\mathbf{1}} \right.$$
$$\left. + \, h_n(L',M;r)\mathbf{Y}^{M}_{L'L'-\mathbf{1}} \right]. \tag{2.30}$$

The coefficients $f_n$, $g_n$ and $h_n$ are derived in Appendix A, so that we merely quote the results here

$$f_n(L',M;r) = \sum_{L=0}^{\infty} R(L;r) \int_0^{2\pi}\int_0^{\pi} \mathbf{Y}^{M*}_{L'L'}(\beta,\phi)\cdot Y_{L0}(\beta)\widehat{\chi}_n \sin\beta d\beta d\phi \tag{2.31}$$

$$= \frac{1}{\sqrt{2}} R(L';r)\left(\delta_{-1M}\widehat{\chi}^*_{-1}\cdot\widehat{\chi}_n - \delta_{1M}\widehat{\chi}^*_{1}\cdot\widehat{\chi}_n\right), \tag{2.32}$$

$$g_n(L',M;r) = \sum_{L=0}^{\infty} R(L;r) \int_0^{2\pi}\int_0^{\pi} \mathbf{Y}^{M*}_{L'L'+1}(\boldsymbol{\beta},\boldsymbol{\phi})\cdot Y_{L0}(\boldsymbol{\beta})\widehat{\chi}_n \sin\boldsymbol{\beta} d\boldsymbol{\beta} d\phi \tag{2.33}$$

$$= R(L'+1;r)\left[\delta_{-1M}\widehat{\chi}^*_{-1}\cdot\widehat{\chi}_n\sqrt{\frac{L'}{2(2L'+3)}}\right.$$
$$\left. + \delta_{1M}\widehat{\chi}^*_{1}\cdot\widehat{\chi}_n\sqrt{\frac{L'}{2(2L'+3)}}\right], \tag{2.34}$$

$$h_n(L',M;r) = \sum_{L=0}^{\infty} R(L;r) \int_0^{2\pi}\int_0^{\pi} \mathbf{Y}^{M*}_{L'L'-1}(\boldsymbol{\beta},\boldsymbol{\phi})\cdot Y_{L0}(\boldsymbol{\beta})\widehat{\chi}_n \sin\boldsymbol{\beta} d\boldsymbol{\beta} d\phi \tag{2.35}$$

$$= R(L'-1;r)\left[\delta_{-1M}\widehat{\chi}^*_{-1}\cdot\widehat{\chi}_n\sqrt{\frac{L'+1}{2(2L'-1)}}\right.$$
$$\left. + \delta_{1M}\widehat{\chi}^*_{1}\cdot\widehat{\chi}_n\sqrt{\frac{L'+1}{2(2L'-1)}}\right]. \tag{2.36}$$

In the following, we write down the explicit form of a plane wave in terms of vector spherical harmonics:

$$\mathbf{E}_k e^{ikr\cos\beta} = \sqrt{2\pi} \sum_{n=-1,1} \left(\mathbf{E}_k \cdot \widehat{\chi}_n^*\right) \sum_{L'} (i)^{L'} \sqrt{(2L'+1)} \left(\widehat{\chi}_{-1}^* \cdot \widehat{\chi}_n \,,\ \widehat{\chi}_1^* \cdot \widehat{\chi}_n\right)$$
$$\cdot \begin{pmatrix} j_{L'}(kr)\, \mathbf{Y}_{L'L'}^{-1} + i\sqrt{\frac{L'}{2L'+1}} j_{L'+1}(kr)\, \mathbf{Y}_{L'L'+1}^{-1} - i\sqrt{\frac{L'+1}{2L'+1}} j_{L'-1}(kr)\, \mathbf{Y}_{L'L'-1}^{-1} \\ -j_{L'}(kr)\, \mathbf{Y}_{L'L'}^{1} + i\sqrt{\frac{L'}{2L'+1}} j_{L'+1}(kr)\, \mathbf{Y}_{L'L'+1}^{1} - i\sqrt{\frac{L'+1}{2L'+1}} j_{L'-1}(kr)\, \mathbf{Y}_{L'L'-1}^{1} \end{pmatrix}. \tag{2.37}$$

This can be further simplified by replacing the $\mathbf{Y}_{L'L'+1}^{\pm 1}$ and $\mathbf{Y}_{L'L'-1}^{\pm 1}$ with taking into account the following relation [9]

$$\frac{i}{k} \nabla \times \left[ j_{L'}(kr)\, \mathbf{Y}_{L'L'}^{M} \right] = \sqrt{\frac{L'}{2L'+1}} j_{L'+1}(kr)\, \mathbf{Y}_{L'L'+1}^{M} - \sqrt{\frac{L'+1}{2L'+1}} j_{L'-1}(kr)\, \mathbf{Y}_{L'L'-1}^{M}. \tag{2.38}$$

Then Eq. (2.37) becomes

$$\mathbf{E}_k e^{ikr\cos\beta} = \sqrt{2\pi} \sum_{n=-1,1} \left(\mathbf{E}_k \cdot \widehat{\chi}_n^*\right) \left(\widehat{\chi}_{-1}^* \cdot \widehat{\chi}_n \,,\ \widehat{\chi}_1^* \cdot \widehat{\chi}_n\right)$$
$$\cdot \begin{pmatrix} \left(1 - \frac{1}{k}\nabla\times\right) \sum_{L'} (i)^{L'} \sqrt{(2L'+1)}\, j_{L'}(kr)\, \mathbf{Y}_{L'L'}^{-\mathbf{1}} \\ \left(-1 - \frac{1}{k}\nabla\times\right) \sum_{L'} (i)^{L'} \sqrt{(2L'+1)}\, j_{L'}(kr)\, \mathbf{Y}_{L'L'}^{\mathbf{1}} \end{pmatrix} \tag{2.39}$$

Finally, we substitute Eq. (2.39) into Eq. (2.16) and use the following relation [7]

$$\mathbf{Y}_{LL}^{M}(\beta,\phi) = \frac{-i(\mathbf{r}\times\nabla) Y_{LM}(\beta,\phi)}{\sqrt{L(L+1)}}, \tag{2.40}$$

together with the definition of the nuclear electric multipole moment [7]:

$$\mathbb{Q}_{LM} = \int_V r^L Y_{LM}^* \,\rho(\mathbf{r}) d^3\mathbf{r} \tag{2.41}$$

and the magnetic multipole moment [8, 9]:

$$\mathbb{M}_{LM} = \frac{1}{c(L+1)} \int_V [\mathbf{r}\times\mathbf{j}(\mathbf{r})] \cdot \nabla \left(r^L Y_{LM}^*\right) d^3\mathbf{r} \tag{2.42}$$

and the definition of the reduced transition probability $\mathbb{B}$

$$\mathbb{B}(\varepsilon L) = \frac{1}{2I_g+1} |\langle I_e \| \widehat{\mathbb{Q}}_L \| I_g \rangle|^2 \tag{2.43}$$

$$\mathbb{B}(\mu L) = \frac{1}{2I_g+1} |\langle I_e \| \widehat{\mathbb{M}}_L \| I_g \rangle|^2. \tag{2.44}$$

We obtain the general formulas for nuclear Rabi frequency and present the main steps in Appendix A, merely quoting here the results for electric/magnetic transitions [8]

$$\langle I_e, M_e|\widehat{H}_I|I_g, M_g\rangle \sim E_k\sqrt{2\pi}\sqrt{\frac{L+1}{L}}\frac{k^{L-1}}{(2L+1)!!}\mathrm{C}_{I_g I_e}(L, M; M_g, M_g) \times \sqrt{2I_g+1}\sqrt{\mathbb{B}(\varepsilon/\mu L)}, \tag{2.45}$$

Here the excited state $|e\rangle$ and the ground state $|g\rangle$ are characterized by the angular momenta $I_e$ and $I_g$, respectively, including their magnetic sublevels $M_e$ and $M_g$. Explicitly, Eq. (2.9) reads

$$\Omega_p(t) = \frac{1}{\hbar}\langle 2|\widehat{H}_I|1\rangle = \frac{4}{\hbar}\sqrt{\frac{\pi I_p(t)}{c\epsilon_0}}\sqrt{\frac{(2I_g+1)(L+1)}{L}}\frac{k_{21}^{L-1}}{(2L+1)!!}\sqrt{\mathbb{B}(\varepsilon/\mu L)}, \tag{2.46}$$

where $I_p(t)$ the intensity of the pump pulse, $L$ the multipolarity of the corresponding nuclear transition and $k_{21}$ the wave number of the corresponding nuclear transition.

Equation (2.46) is used to calculate Rabi frequencies throughout this thesis. We emphasize that the most important parameter $\mathbb{B}(\varepsilon/\mu L)$ characterizing the strength of the nucleus-radiation interaction is obtained from the experimental data, e.g., the Nuclear Structure and Decay Databases [10], such that no first principle calculation involving specific nuclear models is needed.

## 2.2 Maxwell-Bloch Equation

In some experiments, measuring the light signal scattered off a target is the main method to study the interaction between light and matter. In Chaps. 4 and 5 of this thesis, we will encounter such systems, for which the description of only Eq. (2.1) is not complete. To describe the dynamics for both matter and radiation, the coupled Maxwell-Bloch equations[3] must be used [2]:

$$\partial_t\widehat{\rho} = \frac{1}{i\hbar}\left[\widehat{H}, \widehat{\rho}\right] + \widehat{\rho}_s, \tag{2.47}$$

$$\frac{1}{c}\partial_t\Omega + \partial_y\Omega = i\eta\rho_{eg}. \tag{2.48}$$

Equation (2.48) describes the propagation of electromagnetic waves in the forward direction and is derived from Maxwell equations (see Appendix A for the derivation). The backward wave is neglected, because it is not observed in the considered problems. Furthermore, the right hand side of Eq. (2.48) is the source term associated

[3] Also called Maxwell-Schrödinger equations in literature.

with a current or a dipole moment of transition $|e\rangle \leftrightarrow |g\rangle$. The source term $i\eta\rho_{eg}$ will affect the transmission behavior of the incident radiation, where $\eta$ gives the number of nuclei that may scatter the incident photons through the optical path. In Chap. 3, we will solve only Eq. (2.47) since the number of the resonant photons is much greater than the number of nuclei. For the cases treated in Chaps. 4 and 5, the situation is the other way around, the position of Eq. (2.48) becomes important.

### *2.2.1 Coherent Pulse Propagation Through a Resonant Medium*

In this section, we will discuss the system depicted in Fig. 2.1 which is the underlying physical system discussed in for Chaps. 4 and 5. In Fig. 2.1a, a pump pulse propagates through a medium with a length $L$ and interacts with each individual non-mutually interacting nucleus inside the medium as showed in Fig. 2.1a, b. The goal of this example is to calculate the temporal shape of the penetrating pump pulse measured by the detector placed in the forward direction. We consider a two-level nucleus described by a ground state $|1\rangle$ and an excited state $|2\rangle$, and the interaction strength between pump laser and a nucleus is given by $\Omega_p$ with a laser detuning $\Delta_p$ as depicted in Fig. 2.1c.

The theoretical model describing the considered system can be directly obtained from Eqs. (2.6), (2.7), (2.8) and (2.48).

$$\partial_t \rho_{11} = \Gamma\rho_{22} + \frac{i}{2}\left(\Omega_p^*\rho_{21} - \Omega_p\rho_{21}^*\right), \tag{2.49}$$

$$\partial_t \rho_{21} = -\left(\frac{\Gamma}{2} + i\Delta_p\right)\rho_{21} - \frac{i}{2}\Omega_p\left(\rho_{22} - \rho_{11}\right), \tag{2.50}$$

$$\partial_t \rho_{22} = -\Gamma\rho_{22} + \frac{i}{2}\left(\Omega_p\rho_{21}^* - \Omega_p^*\rho_{21}\right), \tag{2.51}$$

and

$$\frac{1}{c}\partial_t\Omega_p + \partial_y\Omega_p = i\eta\rho_{21}. \tag{2.52}$$

The initial and the boundary conditions are:

$$\rho_{eg}(0) = \delta_{1e}\delta_{g1}, \tag{2.53}$$

$$\Omega_p(0, y) = 0, \tag{2.54}$$

$$\Omega_p(t, 0) = \delta(t - \tau), \tag{2.55}$$

where indices $e, g \in \{1, 2\}$. We make here two assumptions: (1) $\Delta_p = 0$ for a resonant pump laser. (2) $\Omega_p \ll \Gamma$ for no Rabi oscillation (i.e., $\Omega_p$ is a perturbation). First, we have to linearize Eqs. (2.49), (2.50) and (2.51). By considering the assumption (2), and substituting

$$\rho_{eg}(t) \rightarrow \delta_{1e}\delta_{g1} + \kappa\rho_{eg}(t), \tag{2.56}$$

$$\Omega_p(t) \rightarrow \kappa\Omega_p(t) \tag{2.57}$$

into Eqs. (2.49)–(2.52), we obtain

$$\kappa\partial_t\rho_{11} = \Gamma\kappa\rho_{22} + \frac{i}{2}\kappa^2\left(\Omega_p^*\rho_{21} - \Omega_p\rho_{21}^*\right), \tag{2.58}$$

$$\kappa\partial_t\rho_{21} = -\frac{\Gamma}{2}\kappa\rho_{21} + \frac{i}{2}\kappa\Omega_p - \frac{i}{2}\kappa^2\Omega_p\left(\rho_{22} - \rho_{11}\right), \tag{2.59}$$

$$\kappa\partial_t\rho_{22} = -\Gamma\kappa\rho_{22} + \frac{i}{2}\kappa^2\left(\Omega_p\rho_{21}^* - \Omega_p^*\rho_{21}\right), \tag{2.60}$$

and

$$\frac{\kappa}{c}\partial_t\Omega_p + \kappa\partial_y\Omega_p = i\eta\kappa\rho_{21}. \tag{2.61}$$

Neglecting the second order $\kappa^2$ terms and subsequently using $\kappa = 1$, we obtain

$$\partial_t\rho_{11} = \Gamma\rho_{22}, \tag{2.62}$$

$$\partial_t\rho_{21} = -\frac{\Gamma}{2}\rho_{21} + \frac{i}{2}\Omega_p, \tag{2.63}$$

$$\partial_t\rho_{22} = -\Gamma\rho_{22}, \tag{2.64}$$

and

$$\frac{1}{c}\partial_t\Omega_p + \partial_y\Omega_p = i\eta\rho_{21}. \tag{2.65}$$

Thus, the dominating equations are

$$\partial_t\rho_{21} = -\frac{\Gamma}{2}\rho_{21} + \frac{i}{2}\Omega_p, \tag{2.66}$$

$$\frac{1}{c}\partial_t\Omega_p + \partial_y\Omega_p = i\eta\rho_{21}. \tag{2.67}$$

By substituting

$$\rho_{21} \rightarrow \Phi e^{-\frac{\Gamma}{2}t}, \tag{2.68}$$

$$\partial_t\rho_{21} \rightarrow -\frac{\Gamma}{2}\Phi e^{-\frac{\Gamma}{2}t} + e^{-\frac{\Gamma}{2}t}\partial_t\Phi, \tag{2.69}$$

$$\Omega_p \rightarrow Ae^{-\frac{\Gamma}{2}t}, \tag{2.70}$$

$$\partial_t\Omega_p \rightarrow -\frac{\Gamma}{2}Ae^{-\frac{\Gamma}{2}t} + e^{-\frac{\Gamma}{2}t}\partial_t A, \tag{2.71}$$

into Eqs. (2.66) and (2.67), we obtain

$$\partial_t \Phi = \frac{i}{2} A, \tag{2.72}$$

$$\frac{1}{c}\left(-\frac{\Gamma}{2} A + \partial_t A\right) + \partial_y A = i\eta\Phi. \tag{2.73}$$

By using the Fourier transform

$$\Phi(t, y) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \phi(\omega, k) e^{i[ky-\omega(t-\tau)]} \tag{2.74}$$

$$A(t, y) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \alpha(\omega, k) e^{i[ky-\omega(t-\tau)]}, \tag{2.75}$$

and substituting

$$\Phi \to \phi, \tag{2.76}$$

$$\partial_t \Phi \to -i\omega\phi, \tag{2.77}$$

$$A \to \alpha, \tag{2.78}$$

$$\partial_t A \to -i\omega\alpha, \tag{2.79}$$

$$\partial_y A \to ik\alpha, \tag{2.80}$$

into Eqs. (2.72) and (2.73), the dispersion relation of the system is obtained

$$k(\omega) = \frac{\omega}{c} - \frac{\eta}{2\omega} - i\frac{\Gamma}{2c}. \tag{2.81}$$

By using the inverse Fourier transform, the solution of $\alpha$ is obtained

$$A\,(t, y) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \alpha_0 e^{-i[k(\omega)y-\omega(t-\tau)]} d\omega \tag{2.82}$$

$$= \frac{1}{\sqrt{2\pi}} e^{-\frac{\Gamma}{2c}y} \int_{-\infty}^{\infty} \alpha_0 e^{-i\left[\left(\frac{\omega}{c}-\frac{\eta}{2\omega}\right)y-\omega(t-\tau)\right]} d\omega. \tag{2.83}$$

Here, $\alpha_0 = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \delta(t-\tau) e^{-i\omega(t-\tau)} dt = \frac{1}{\sqrt{2\pi}}$ is the Fourier transform of the boundary condition (2.55). Finally, the solution of $\Omega_p$ is (complete derivation is presented in Appendix A)

$$\Omega_p(t, y) = \frac{1}{2\pi} e^{-\frac{\Gamma}{2}\left[\frac{y}{c}+(t-\tau)\right]} \int_{-\infty}^{\infty} e^{-i\left[\left(\frac{\omega}{c}-\frac{\eta}{2\omega}\right)y-\omega(t-\tau)\right]} d\omega \tag{2.84}$$

$$= \frac{1}{2\pi} e^{-\frac{\Gamma}{2}[\frac{y}{c}+\beta]} \sum_{n=0}^{\infty} \frac{(iq)^n}{n!} \int_{-\infty}^{\infty} \frac{1}{\omega^n} e^{-i\omega z} d\omega \tag{2.85}$$

$$= \frac{1}{\sqrt{2\pi}} e^{-\frac{\Gamma}{2}[\frac{y}{c}+\beta]} \sum_{n=0}^{\infty} \frac{(iq)^n}{n!} \left[ -i\sqrt{\frac{\pi}{2}} \frac{(-iz)^{n-1}}{(n-1)!} sgn(z) \right] \tag{2.86}$$

$$= \delta(z) e^{-\frac{\Gamma}{2}[\frac{y}{c}+\beta]} - q \frac{J_1\left(2\sqrt{qz'}\right)}{2\sqrt{qz'}} e^{-\frac{\Gamma}{2}[\frac{y}{c}+\beta]}. \tag{2.87}$$

Here, $J_1(z)$ is the Bessel function of first kind [11, 12], $\beta = t - \tau$, $z = \frac{y}{c} - (t - \tau)$, $z' = -z$ and $sgn(z)$ is the sign function [13] which equals -1 in our case since $z < 0$. Additionally, in most cases $\frac{L}{c}$ is much smaller than $t - \tau$, i.e., the prorogation time of the incident light pulse through a target with length $L$ is much shorter than the detection time window $t - \tau$. Therefore, the terms $\frac{y}{c}$ are typically negligible. Moreover, we derive Eq. (2.86) from Eq. (2.85) by using the Fourier transform of $1/\omega^n$. The explicit form of Eq. (2.87) is then

$$\begin{aligned} \Omega_p(t, y) &= \left\{ \delta\left[\frac{y}{c} - (t-\tau)\right] - \left(\frac{\eta y}{2}\right) \frac{J_1\left[2\sqrt{\left(\frac{\eta y}{2}\right)\left(t - \tau - \frac{y}{c}\right)}\right]}{2\sqrt{\left(\frac{\eta y}{2}\right)\left(t - \tau - \frac{y}{c}\right)}} \right\} e^{-\frac{\Gamma}{2}\left(\frac{y}{c}+t-\tau\right)} \\ &= \left\{ \delta\left[\frac{y}{c} - (t-\tau)\right] - \left(\frac{\xi\Gamma y}{4L}\right) \frac{J_1\left[2\sqrt{\left(\frac{\xi\Gamma y}{4L}\right)\left(t - \tau - \frac{y}{c}\right)}\right]}{2\sqrt{\left(\frac{\xi\Gamma y}{4L}\right)\left(t - \tau - \frac{y}{c}\right)}} \right\} e^{-\frac{\Gamma}{2}\left(\frac{y}{c}+t-\tau\right)}. \end{aligned} \tag{2.88}$$

In experiments, the measured field [1, 11, 12, 14] is proportional to

$$\Omega_p(t, L) = \left\{ \delta\left[\frac{L}{c} - (t-\tau)\right] - \left(\frac{\xi\Gamma}{4}\right) \frac{J_1\left[2\sqrt{\left(\frac{\xi\Gamma}{4}\right)\left(t - \tau - \frac{L}{c}\right)}\right]}{2\sqrt{\left(\frac{\xi\Gamma}{4}\right)\left(t - \tau - \frac{L}{c}\right)}} \right\} e^{-\frac{\Gamma}{2}\left(\frac{L}{c}+t-\tau\right)}, \tag{2.89}$$

and the measured signal is proportional to

$$|\Omega_p(t, L)|^2 = \left\{ \delta\left[\frac{L}{c} - (t-\tau)\right] - \left(\frac{\xi\Gamma}{4}\right) \frac{J_1\left[2\sqrt{\left(\frac{\xi\Gamma}{4}\right)\left(t - \tau - \frac{L}{c}\right)}\right]}{2\sqrt{\left(\frac{\xi\Gamma}{4}\right)\left(t - \tau - \frac{L}{c}\right)}} \right\}^2 e^{-\Gamma\left(\frac{L}{c}+t-\tau\right)}. \tag{2.90}$$

This equation can be derived also using an iterative method with the response function as it was shown in Ref. [15]. We will use Eq. (2.90) in Chaps. 4 and 5 to explain a phenomenon called dynamical beat [16] of nuclear forward scattering [1].

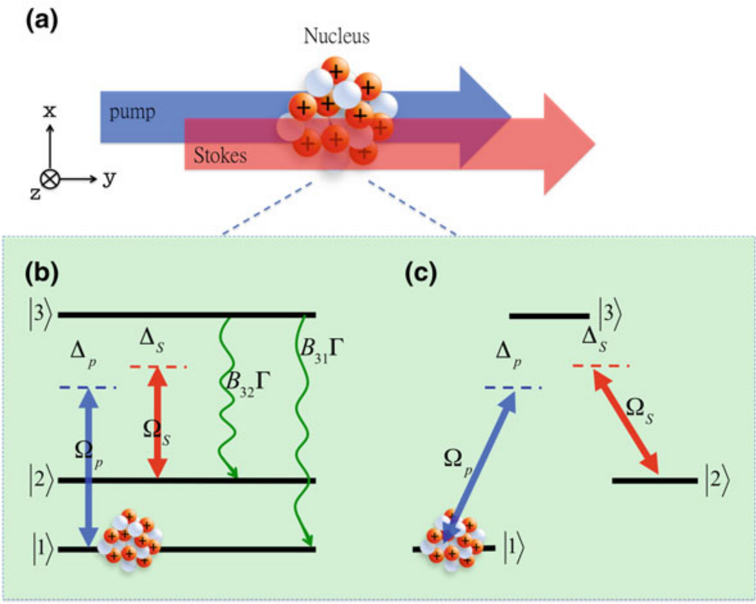


**Fig. 2.2** **a** Interaction between a nucleus, pump laser (*blue wide arrow*) and Stokes laser (*red wide arrow*). Typical sketches of a three-level Λ-type system appear in the literature of **b** nuclear and **c** atomic physics. The *blue* ($\Omega_p$) and *red* ($\Omega_S$) *vertical arrows* depict pump and Stokes lasers, respectively. $\Delta_p$ and $\Delta_S$ denote the detunings of the corresponding lasers. The *green wiggled arrows* illustrate the spontaneous decay of state $|3\rangle$, $B_{31}$ ($B_{32}$) is the branching ratio of $|3\rangle \leftrightarrow |1\rangle$ ($|3\rangle \leftrightarrow |2\rangle$) transition and $\Gamma$ is the spontaneous decay rate of state $|3\rangle$

## 2.3 Three-Level Λ-Type System

In this section, we extend the used theory for a three-level Λ-type system [2] that is well known for describing several effects in atomic quantum optics, for example, stimulated Raman adiabatic passage (STIRAP) [3] and electromagnetically induced transparency (EIT) [4–6]. A typical three-level Λ-type system is illustrated in Fig. 2.2a. For convenience, we assume the wave function of the considered nucleus is $|\psi\rangle = C_1(t)|1\rangle + C_2(t)|2\rangle + C_3(t)|3\rangle$. The interaction between the nucleus and two lasers is typically illustrated by a sketch like Fig. 2.2b in nuclear physics or Fig. 2.2c in atomic physics. Since the pattern of Fig. 2.2c looks like the Greek letter Λ, it is called as a Λ-type system. Here, pump laser (blue arrow) drives the transition $|3\rangle \leftrightarrow |1\rangle$, with detuning $\Delta_p$ and Stokes laser (red arrow) drives the transition $|3\rangle \leftrightarrow |2\rangle$ with detuning $\Delta_S$. The explicit form of $\widehat{\rho}$ in this case is:

$$\widehat{\rho} = \begin{pmatrix} \rho_{11} & \rho_{12} & \rho_{13} \\ \rho_{21} & \rho_{22} & \rho_{23} \\ \rho_{31} & \rho_{32} & \rho_{33} \end{pmatrix}. \tag{2.91}$$

By considering the spontaneous decay, the decoherence matrix is

$$\widehat{\rho_s} = \frac{\Gamma}{2}\begin{pmatrix} 2B_{31}\rho_{33} & 0 & -\rho_{13} \\ 0 & 2B_{32}\rho_{33} & -\rho_{23} \\ -\rho_{31} & -\rho_{32} & -2\rho_{33} \end{pmatrix}. \tag{2.92}$$

In Eq. (2.92), $B_{31} + B_{32} = 1$ due to $\rho_{11} + \rho_{22} + \rho_{33} = 1$, i.e., the conservation of population in a closed three level system. The decay rate $\Gamma/2$ for each of diagonal coherence is derived from Eq. (2.3). Without any external laser, $\rho_{33}(t) \sim e^{-\Gamma t}$ which means $C_3(t) \sim e^{-(\Gamma/2)t}$, whence the other coherence $\rho_{3\mu}$ and $\rho_{\mu 3}$ are also proportional to $e^{-(\Gamma/2)t}$. On the other hand, $\widehat{\rho_{s}}_{,12} = \widehat{\rho_{s}}_{,21} = 0$ as the two lower states $|1\rangle$ and $|2\rangle$ do not experience any decoherence process in this example. This corresponds to choosing an isomer state $|2\rangle$, whose decay is strongly hindered.

The interaction Hamiltonian matrix $\widehat{H}(t)$ is [2, 3]:

$$\widehat{H} = -\frac{\hbar}{2}\begin{pmatrix} 0 & 0 & \Omega_p^* \\ 0 & -2\left(\triangle_p - \triangle_S\right) & \Omega_S^* \\ \Omega_p & \Omega_S & 2\triangle_p \end{pmatrix}, \tag{2.93}$$

where $\hbar$ is the reduced Planck constant. The explicit form of Eq. (2.1) can be obtained by substituting Eqs. (2.91), (2.92) and (2.93) into Eq. (2.1):

$$\partial_t\rho_{11} = B_{31}\Gamma\rho_{33} + \frac{i}{2}\left(\Omega_p^*\rho_{31} - \Omega_p\rho_{31}^*\right) \tag{2.94}$$

$$\partial_t\rho_{21} = i\left(\Delta_S - \Delta_p\right)\rho_{21} + \frac{i}{2}\left(\Omega_S^*\rho_{31} - \Omega_p\rho_{32}^*\right) \tag{2.95}$$

$$\partial_t\rho_{31} = -\left(\frac{\Gamma}{2} + i\Delta_p\right)\rho_{31} + \frac{i}{2}\Omega_S\rho_{21} - \frac{i}{2}\Omega_p\left(\rho_{33} - \rho_{11}\right) \tag{2.96}$$

$$\partial_t\rho_{22} = B_{32}\Gamma\rho_{33} + \frac{i}{2}\left(\Omega_S^*\rho_{32} - \Omega_S\rho_{32}^*\right) \tag{2.97}$$

$$\partial_t\rho_{32} = -\left(\frac{\Gamma}{2} + i\Delta_S\right)\rho_{32} + \frac{i}{2}\Omega_p\rho_{21}^* - \frac{i}{2}\Omega_S\left(\rho_{33} - \rho_{22}\right) \tag{2.98}$$

$$\partial_t\rho_{33} = -\Gamma\rho_{33} + \frac{i}{2}\left(\Omega_p\rho_{31}^* - \Omega_p^*\rho_{31}\right) + \frac{i}{2}\left(\Omega_S\rho_{32}^* - \Omega_S^*\rho_{32}\right). \tag{2.99}$$

In above equations, $\Omega_p$ and $\Omega_S$ are Rabi frequencies defined as:

$$\begin{aligned}\Omega_p(t) &= \frac{1}{\hbar}\langle 3|\widehat{H}_I|1\rangle \\ &= \frac{4}{\hbar}\sqrt{\frac{\pi I_p(t)}{c\epsilon_0}}\sqrt{\frac{(2I_1+1)(L_{31}+1)}{L_{13}}}\frac{k_{31}^{L_{31}-1}}{(2L_{13}+1)!!}\sqrt{\mathbb{B}(\varepsilon/\mu L_{13})}, \qquad (2.100)\\ \Omega_S(t) &= \frac{1}{\hbar}\langle 3|\widehat{H}_I|2\rangle\end{aligned}$$

$$= \frac{4}{\hbar}\sqrt{\frac{\pi I_S(t)}{c\epsilon_0}}\sqrt{\frac{(2I_2+1)(L_{23}+1)}{L_{23}}}\frac{k_{32}^{L_{23}-1}}{(2L_{23}+1)!!}\sqrt{\mathbb{B}(\varepsilon/\mu\, L_{23})}, \quad (2.101)$$

where $I_{p(S)}(t)$ is the pump (Stokes) pulse, $I_{1(2)}$ is the angular momentum of ground state $|1(2)\rangle$, and , $L_{1(2)3}$ is the multipolarity of the corresponding nuclear transition $|1(2)\rangle \leftrightarrow |3\rangle$. Equations (2.94)–(2.101) are successfully used to explain plenty of phenomena in atomic quantum optics. In this thesis, we adopt and use it to investigate the proposal called nuclear coherent population transfer (NCPT) in Chap. 3. Furthermore, Eqs. (2.94)–(2.101) together with

$$\frac{1}{c}\partial_t\Omega_p + \partial_y\Omega_p = i\eta\rho_{31}, \quad (2.102)$$

will be used to discuss another proposal labeled as electromagnetically modified nuclear forward scattering in Chap. 5.

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