

Chapter 2

Theoretical Background

In this chapter some of the fundamental theoretical tools, required to characterize the first excited isomeric state of ^{229}Th , are developed. The first section provides a short introduction to nuclear shell models with a focus on the Nilsson single-particle shell model and rotational bands. The second section describes the fundamental theories of nuclear γ decay and internal conversion, together with their applications to $^{229\text{m}}\text{Th}$. The last section introduces the theoretical background for direct nuclear laser coupling and gives examples for $^{229\text{m}}\text{Th}$. Also $^{235\text{m}}\text{U}$ is discussed for comparison, being the isomeric state of second lowest excitation energy.

2.1 Nuclear Shell Models

The ultimate way to describe nuclear transitions would be within a quantum chromodynamical framework. Unfortunately, this theory has not yet been developed to a point which would allow for precise predictions of level schemes. Especially complex nuclei and transitions of lowest energies, as is the case for the first excited state of ^{229}Th , is far beyond the scope of quantum chromodynamics at the current time. Therefore, all possible predictions are within the framework of different collective models, of which the most important one is the nuclear shell model. This model, even with all possible extensions, still does not allow for a precise prediction of nuclear transitions in the eV-range. Instead, it allows for rough estimations, which can then be compared to experimental data. In the following, the nuclear shell model will be discussed together with its extensions as far as of interest in the context of this thesis. The discussion is based on Ref. [1].

2.1.1 Nuclear Single Particle Shell Model

Similar to the electrons in an atom, the nucleons can be described as solutions of the Schrödinger equation

$$\hat{H}\psi = E\psi \quad (2.1)$$

for a given Hamiltonian $\hat{H}$. Assuming that a potential is found, in which each nucleon moves independently of all others, the total Hamiltonian can be written as a sum over individual particle Hamiltonians [1]

$$\hat{H} = \sum_i \hat{H}_i = \sum_i \left[-\frac{\hbar^2}{2m} \Delta_i + V(r_i) \right], \quad (2.2)$$

where $V(r)$ is the potential of interest. The Schrödinger equation must then hold for each individual particle

$$\hat{H}_i \psi_i = \left[-\frac{\hbar^2}{2m} \Delta_i + V(r_i) \right] \psi_i = E_i \psi_i. \quad (2.3)$$

The potential in which one nucleon is moving is given by the mean field that is generated by all other nucleons and therefore corresponds to the nucleon density, which is described by Fermi statistics. The basic potential approach is therefore the Woods–Saxon potential

$$V_{\text{Woods–Saxon}}(r) = \frac{-V_0}{1 + e^{(r-R)/a}}, \quad (2.4)$$

where V_0 corresponds to the potential depth, R is the radius at which the potential depth is half of V_0 and a is a parameter corresponding to the steepness of the potential. V_0 , R and a have to be experimentally identified. A comparison of theoretical predictions from this model and measured level schemes reveals that this simple approach does not lead to satisfying predictions. Instead also the spin-orbit interaction has to be considered. This is done via an extra term in the potential, which then becomes

$$V(r) = \frac{-V_0}{1 + e^{(r-R)/a}} + V_{ls}(r) \frac{\langle \hat{l}\hat{s} \rangle}{\hbar^2}. \quad (2.5)$$

Here V_{ls} is a scaling potential and $\hat{l}$ and $\hat{s}$ are the orbital momentum and spin operator, respectively. The operators are coupled as $\hat{j} = \hat{l} + \hat{s}$ which, by applying the commutativity of $\hat{l}$ and $\hat{s}$, gives

$$\hat{l}\hat{s} = \frac{1}{2} \left(\hat{j}^2 - \hat{l}^2 - \hat{s}^2 \right). \quad (2.6)$$

As all components of $\hat{j}$ commute with the Hamiltonian, one can find a basis of states $|slj\rangle$, for which the eigenvalue relations of the angular momentum operator hold

$$\begin{aligned}\hat{s}^2|slj\rangle &= \hbar^2 s(s+1)|slj\rangle, \\ \hat{l}^2|slj\rangle &= \hbar^2 l(l+1)|slj\rangle, \\ \hat{j}^2|slj\rangle &= \hbar^2 j(j+1)|slj\rangle.\end{aligned}\tag{2.7}$$

Thus the expectation value $\langle \hat{l}\hat{s} \rangle$ becomes

$$\langle \hat{l}\hat{s} \rangle = \frac{\hbar^2 [j(j+1) - l(l+1) - s(s+1)]}{2}.\tag{2.8}$$

Inserting this relation into Eq. (2.5) leads to

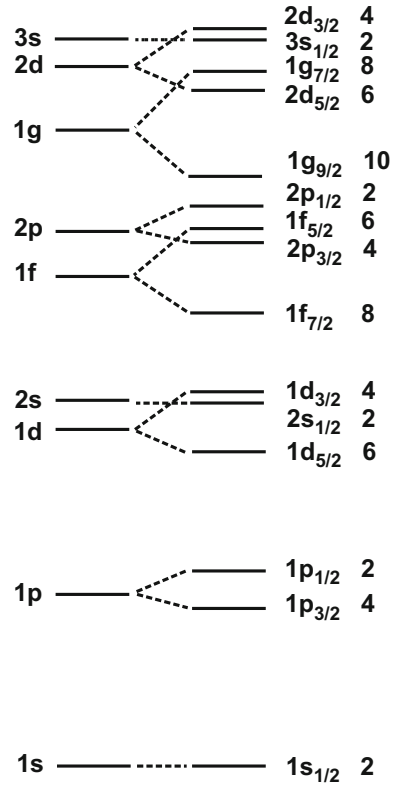
$$V(r) = \frac{-V_0}{1 + e^{(r-R)/a}} + V_{ls}(r) \frac{j(j+1) - l(l+1) - s(s+1)}{2}.\tag{2.9}$$

It is known from experiment that $V_{ls}(r)$ takes negative values. The resulting Schrödinger equation cannot be solved analytically, but due to the spherical symmetry of the potential the resulting wave functions can be factorized into a radial wave function $R_{nl}(r)$ and the spherical harmonics $Y_{lm}(\theta, \phi)$, where n , l and m are the radial quantum number, the orbital quantum number and the magnetic quantum number, which can take values from 1 to ∞ , 0 to ∞ and $-l$ to l respectively. The spin quantum number s is involved to also take spin-orbital coupling into account. The spin s takes the values $-1/2$ or $+1/2$. It turns out that the energy is independent of the magnetic quantum number m . An energy eigenstate is therefore fixed by the three quantum numbers n , l and s . Usually the states are assigned by n , l and $j = l + s$ instead, where for l the letters s, p, d, f, g, h,... (corresponding to $l = 0, 1, 2, \dots$) are used. Due to the independence on m , each state is $(2j + 1)$ times degenerate. The numerically calculated level schemes for the Woods–Saxon potential with and without spin-orbital coupling are shown in Fig. 2.1.

2.1.2 The Nilsson Model

The above made assumption of a spherical nuclear potential cannot be validated for all nuclei. In general, deformation occurs, leading to changes of the energy eigenvalues, which have to be considered in a more realistic approach. This is especially important for nuclei far from closed shells, as is the case for ^{229}Th . Therefore deformations of the Woods–Saxon potential have to be allowed, which again can only be done numerically. In order to get any insights into the theory of deformed nuclei, the deformed harmonic oscillator potential is considered in the following. This is also the way in which deformation was first introduced into the model by Nilsson [2].

Fig. 2.1 Energy levels of the nuclear single-particle shell model with (right) and without (left) spin-orbital coupling. The corresponding quantum numbers are assigned to each state. The degeneracy of the states are given on the right according to $2j + 1$



The Hamiltonian of the anisotropic harmonic oscillator is of the form [1]

$$\hat{H}_0 = -\frac{\hbar^2}{2m} \Delta + \frac{m}{2} (\omega_x^2 x^2 + \omega_y^2 y^2 + \omega_z^2 z^2). \quad (2.10)$$

The energy eigenvalues of this Hamiltonian are

$$E_0 = \hbar\omega_x \left(n_x + \frac{1}{2} \right) + \hbar\omega_y \left(n_y + \frac{1}{2} \right) + \hbar\omega_z \left(n_z + \frac{1}{2} \right), \quad (2.11)$$

where n_x , n_y and n_z denote the quantum numbers characterizing the corresponding eigenstates. For the following, axial symmetry with $\omega_x = \omega_y$ is assumed. Further, a constant potential volume $\omega_x \omega_y \omega_z = \text{const.} = \omega_0^3$ is introduced and a deformation parameter δ is introduced as

$$\delta = 2 \cdot \frac{b - a}{b + a}, \quad (2.12)$$

where b denotes the length of the ellipsoid along the symmetry axis and a the length orthogonal to the symmetry axis. Thus negative values of δ correspond to oblate deformations and positive values to prolate deformations. With the help of this deformation parameter, ω_x , ω_y as well as ω_z are expressed as

$$\begin{aligned}\omega_x^2 &= \omega_y^2 = \omega^2(\delta) \left(1 + \frac{2}{3}\delta\right) \\ \omega_z^2 &= \omega^2(\delta) \left(1 - \frac{4}{3}\delta\right).\end{aligned}\tag{2.13}$$

In this case one has

$$\omega_x \omega_y \omega_z = \omega^3(\delta) \left(1 - \frac{4}{3}\delta^2 - \frac{16}{27}\delta^3\right)^{1/2} = \omega_0^3,\tag{2.14}$$

and correspondingly

$$\omega(\delta) = \omega_0 \left(1 - \frac{4}{3}\delta^2 - \frac{16}{27}\delta^3\right)^{-1/6}.\tag{2.15}$$

In the original work [2], Nilsson introduces a deformation dependent oscillator length $b(\delta) = (\hbar/m\omega_0(\delta))^{1/2}$ and dimensionless coordinates $\vec{r}' = \vec{r}/b$ to transform the Hamiltonian of Eq. (2.10) to

$$\hat{H}_0 = \hbar\omega(\delta) \left(-\frac{1}{2}\Delta' + \frac{r'^2}{2} - \frac{4}{3}\sqrt{\frac{\pi}{5}}\delta r'^2 Y_{20}(\theta', \phi')\right).\tag{2.16}$$

It is convenient to transform to cylindrical coordinates, in which case the eigenstates are characterized by the quantum numbers n_z , n_ρ and m_l , where m_l denotes the projection of the orbital angular momentum onto the symmetry axis and is often denoted as Λ . Correspondingly one obtains

$$\begin{aligned}E_0 &= \hbar\omega_z \left(n_z + \frac{1}{2}\right) + \hbar\omega_{xy} (2n_\rho + m_l + 1) \\ &= \hbar\omega_0 \left[\left(N + \frac{3}{2}\right) + \delta \left(\frac{N}{3} - n_z\right)\right].\end{aligned}\tag{2.17}$$

Here $N = n_x + n_y + n_z$ was introduced as a new quantum number and $\omega_{xy} = \omega_x = \omega_y$. The axial symmetry of the Hamiltonian causes Λ to be a good quantum number. The same is valid for the z-component of the spin Σ and the z-component of the total angular momentum $\Omega = \Lambda + \Sigma$. For this reason, the eigenstates of the given Hamiltonian are described by the quantum numbers

$(2j + 1)/2$ substates during deformation. After introduction of the correction terms $C < \hat{l}\hat{s} >$ and $D\hat{l}^2$, Ω and π are the only remaining good quantum numbers of the states. However, for large deformations, the $\hat{l}\hat{s}$ and $\hat{l}^2$ term are less important and the quantum numbers in Eq. (2.18) are approximately good quantum numbers. For this reason they are denoted as asymptotic quantum numbers and used to describe the Nilsson states.

In case of ^{229}Th , the nuclear ground state is described by the Nilsson quantum numbers $5/2^+633$ and the isomeric state by $3/2^+631$. The deformation parameter is often defined as ϵ_2 , the Nilsson quadrupole deformation parameter. ϵ_2 relates to δ as

$$\epsilon_2 = \delta + \frac{1}{6}\delta^2 + \frac{5}{18}\delta^3 + \dots \quad (2.20)$$

For ^{229}Th , the nuclear deformation can be estimated from the electric quadrupole moment to $\epsilon_2 \approx 0.19$ [4]. This is in good agreement with the crossing point of the $5/2^+633$ ground state and the $3/2^+631$ isomeric state in the Nilsson diagram, as shown as a red circle in Fig. 2.2.

2.1.3 Rotational Bands

In order to understand the nuclear γ -ray spectrum of ^{229}Th , also rotational motion of the nucleus has to be considered. As the nucleus is deformed, rotations around an axis perpendicular to the symmetry axis of the nucleus can occur. In case of cylindrical symmetry, the corresponding Hamilton operator takes the form [1]

$$\hat{H}_{\text{rot}} = \frac{\hat{I}^2 - \hat{I}_z^2}{2\theta_x} + \frac{\hat{I}_z^2}{\theta_z}. \quad (2.21)$$

Here $\hat{I}$ denotes the total angular momentum operator of the rotation, and $\hat{I}_z$ is the corresponding projection onto the z -axis of the body-fixed system. θ_x and θ_z denote the moments of inertia in x and z direction, respectively. Introducing the new quantum number $K = I_z$ as the z -component of the total angular momentum, the rotational energy of the system can be obtained by solving the Schrödinger equation with the given Hamiltonian as

$$E_{\text{rot}} = \frac{\hbar^2}{2\theta_x} (I(I + 1) - K^2) + \frac{\hbar^2}{2\theta_z} K^2. \quad (2.22)$$

Based on symmetry arguments, it is possible to show that in case of axial and reflection symmetry $K = 0$ and $I = 0, 2, 4, \dots$ holds and the rotational energy is given by [1]

$$E_{\text{rot}} = \frac{\hbar^2}{2\theta_x} (I(I + 1)). \quad (2.23)$$

In accordance with Eq. (2.23), there is a series of excited rotational states, called rotational band, corresponding to each energy level of the Nilsson model. The ground state of each rotational band is denoted as the band head. Besides rotational states also vibrational states can play a role. These will not be discussed, as they are not of importance in the considered context.

2.2 Nuclear γ Decay and Internal Conversion

Two decay channels of $^{229\text{m}}\text{Th}$ are addressed in this work. These are the γ decay via emission of a photon in the vacuum ultra-violet (VUV) region around 160 nm and the internal conversion (IC) decay under emission of a low-energy electron. The search for an internal conversion decay channel has led to the successful observation of the isomeric decay (see Sect. 5.2.2). In the following, the theoretical background is discussed for both decay channels.

2.2.1 Nuclear γ Decay

The theory of nuclear coupling to electromagnetic radiation provides the basis for nuclear γ decay as well as excitation. In the following, the γ -emission rate for a given nuclear transition will be derived. For this purpose, the electromagnetic field is developed in multipoles, closely following the procedure described in Ref. [5]. The starting point are the Maxwell equations, which take the following form (in SI units):

$$\begin{aligned} \vec{\nabla} \cdot \vec{E}(\vec{r}, t) &= \frac{\rho(\vec{r}, t)}{\epsilon_0} \\ \vec{\nabla} \cdot \vec{H}(\vec{r}, t) &= -\vec{\nabla} \cdot \vec{M}(\vec{r}, t) \\ \vec{\nabla} \times \vec{E}(\vec{r}, t) &= -\mu_0 \frac{\partial(\vec{H}(\vec{r}, t) + \vec{M}(\vec{r}, t))}{\partial t} \\ \vec{\nabla} \times \vec{H}(\vec{r}, t) &= \epsilon_0 \frac{\partial \vec{E}(\vec{r}, t)}{\partial t} + \vec{J}(\vec{r}, t). \end{aligned} \quad (2.24)$$

These are the usual vacuum Maxwell equations after introduction of a magnetization $\vec{M}(\vec{r}, t)$, in order to take the intrinsic angular momenta of the particles into account. $\vec{E}(\vec{r}, t)$ denotes the electric field, $\vec{H}(\vec{r}, t)$ is the magnetic field and $\vec{J}(\vec{r}, t)$ as well as $\rho(\vec{r}, t)$ denote the current and charge density, respectively. ϵ_0 and μ_0 are the vacuum permittivity and permeability constants.

Assuming a single source frequency ω , the time dependency can be separated as [6]

$$\begin{aligned}\rho(\vec{r}, t) &= \rho(\vec{r})e^{-i\omega t} + \rho^*(\vec{r})e^{i\omega t} \\ \vec{J}(\vec{r}, t) &= \vec{J}(\vec{r})e^{-i\omega t} + \vec{J}^*(\vec{r})e^{i\omega t} \\ \vec{M}(\vec{r}, t) &= \vec{M}(\vec{r})e^{-i\omega t} + \vec{M}^*(\vec{r})e^{i\omega t}.\end{aligned}\tag{2.25}$$

The same relation then also holds for the electric and magnetic fields. By inserting these relations into Eq. (2.24) and applying the continuity equation

$$\frac{\partial \rho(\vec{r}, t)}{\partial t} + \vec{\nabla} \cdot \vec{J}(\vec{r}, t) = 0,\tag{2.26}$$

the Maxwell equations take the following form

$$\begin{aligned}\vec{\nabla} \cdot \vec{E}(\vec{r}) &= -\frac{i}{k} \sqrt{\frac{\mu_0}{\epsilon_0}} \vec{\nabla} \cdot \vec{J}(\vec{r}) \\ \vec{\nabla} \cdot \vec{H}(\vec{r}) &= -\vec{\nabla} \cdot \vec{M}(\vec{r}) \\ \vec{\nabla} \times \vec{E}(\vec{r}) &= ik \sqrt{\frac{\mu_0}{\epsilon_0}} \left(\vec{H}(\vec{r}) + \vec{M}(\vec{r}) \right) \\ \vec{\nabla} \times \vec{H}(\vec{r}) &= -ik \sqrt{\frac{\epsilon_0}{\mu_0}} \vec{E}(\vec{r}) + \vec{J}(\vec{r}).\end{aligned}\tag{2.27}$$

Here $\omega = c \cdot k$ with k as the wavenumber and $c = 1/\sqrt{\epsilon_0 \mu_0}$ was used. Applying the curl operation to the last two equations and making use of the identity

$$\vec{\nabla} \times (\vec{\nabla} \times \vec{V}) = \vec{\nabla}(\vec{\nabla} \cdot \vec{V}) - \vec{\nabla}^2 \vec{V},\tag{2.28}$$

the inhomogeneous Helmholtz equations are derived

$$\begin{aligned}(\vec{\nabla}^2 + k^2) \vec{E}(\vec{r}) &= -ik \sqrt{\frac{\mu_0}{\epsilon_0}} \left(\vec{J}(\vec{r}) + \vec{\nabla} \times \vec{M}(\vec{r}) \right) - \frac{i}{k} \sqrt{\frac{\mu_0}{\epsilon_0}} \vec{\nabla}(\vec{\nabla} \cdot \vec{J}(\vec{r})) \\ (\vec{\nabla}^2 + k^2) \vec{H}(\vec{r}) &= -k^2 \vec{M}(\vec{r}) - \vec{\nabla} \times \vec{J}(\vec{r}) - \vec{\nabla}(\vec{\nabla} \cdot \vec{M}(\vec{r})).\end{aligned}\tag{2.29}$$

These equations take the form of an inhomogeneous wave equation

$$(\vec{\nabla}^2 + k^2) \vec{\Psi}(\vec{r}) = -\vec{V}(\vec{r}),\tag{2.30}$$

which is solved with the help of the Green's formalism. The solution for the vector component α reads

$$\Psi_\alpha(\vec{r}) = \sum_\beta \int d\vec{r}' G_{\alpha\beta}(\vec{r}, \vec{r}') V_\beta(\vec{r}'),\tag{2.31}$$

where the Green's function $G_{\alpha\beta}$ is expressed in terms of vector spherical harmonics as [5]

$$G_{\alpha\beta}(\vec{r}, \vec{r}') = \sum_{l=0}^{\infty} \sum_{m=-l}^l \frac{ik}{\sqrt{l(l+1)}} h_l^{(1)}(kr) j_l(kr') X_{lm\alpha} X_{lm\beta}^*(\theta', \phi'). \quad (2.32)$$

Here $h_l^{(1)}$ denote the spherical Hankel functions and j_l the spherical Bessel functions. $\vec{X}_{lm}(\theta, \phi) = \vec{L} Y_{lm}(\theta, \phi) / \sqrt{l(l+1)}$ denote the vector spherical harmonics, as developed from the scalar spherical harmonics $Y_{lm}(\theta, \phi)$ with help of the angular momentum operator $\vec{L} = -i\vec{r} \times \vec{\nabla}$. In this way $\vec{X}_{lm}^* = Y_{lm}^* \vec{L} / \sqrt{l(l+1)}$ becomes a differential operator that acts on the inhomogeneity $\vec{V}$. The solution to the inhomogeneous wave equation (2.30) thus reads

$$\vec{\Psi}(\vec{r}) = \sum_{l=0}^{\infty} \sum_{m=-l}^l h_l^{(1)}(kr) \vec{X}_{lm}(\theta, \phi) \frac{ik}{\sqrt{l(l+1)}} \int d\vec{r}' j_l(kr') Y_{lm}^*(\theta', \phi') \vec{L}' \cdot \vec{V}(\vec{r}'). \quad (2.33)$$

This general solution can be used to solve the inhomogeneous Helmholtz equations (2.29), which will be done in the following only for the first equation with the special inhomogeneity

$$\vec{V}(\vec{r}) = ik \sqrt{\frac{\mu_0}{\epsilon_0}} \left(\vec{J}(\vec{r}) + \vec{\nabla} \times \vec{M}(\vec{r}) \right) + \frac{i}{k} \sqrt{\frac{\mu_0}{\epsilon_0}} \vec{\nabla} (\vec{\nabla} \cdot \vec{J}(\vec{r})). \quad (2.34)$$

The solution for the electric field is determined to

$$\vec{E}(\vec{r}) = \sum_{l=0}^{\infty} \sum_{m=-l}^l a_{lm}^{(M)} h_l^{(1)}(kr) \vec{X}_{lm}(\theta, \phi), \quad (2.35)$$

with the field amplitude of order l, m

$$a_{lm}^{(M)} = \frac{ik}{\sqrt{l(l+1)}} \int d^3r' j_l(kr') Y_{lm}^*(\theta', \phi') \vec{L}' \cdot \left[ik \sqrt{\frac{\mu_0}{\epsilon_0}} \left(\vec{J}(\vec{r}') + \vec{\nabla} \times \vec{M}(\vec{r}') \right) + \frac{i}{k} \sqrt{\frac{\mu_0}{\epsilon_0}} \vec{\nabla} (\vec{\nabla} \cdot \vec{J}(\vec{r}')) \right]. \quad (2.36)$$

Making use of the identities

$$\begin{aligned} \vec{L} \cdot \vec{V}(\vec{r}) &= i \vec{\nabla} \cdot (\vec{r} \times \vec{V}(\vec{r})), \\ \vec{L} \cdot (\vec{\nabla} \times \vec{V}(\vec{r})) &= i \vec{\nabla}^2 (\vec{r} \cdot \vec{V}(\vec{r})) - \frac{i \partial(r^2 \vec{\nabla} \cdot \vec{V}(\vec{r}))}{r \partial r}, \text{ and} \\ \vec{L} \cdot \vec{\nabla}_S(\vec{r}) &= 0, \end{aligned} \quad (2.37)$$

this equation is simplified to

$$a_{lm}^{(M)} = \frac{-ik^2}{\sqrt{l(l+1)}} \sqrt{\frac{\mu_0}{\epsilon_0}} \int d\vec{r}' j_l(kr') Y_{lm}^*(\theta', \phi') \cdot \left[\vec{\nabla} \cdot (\vec{r}' \times \vec{J}(\vec{r}')) + \vec{\nabla}^2 (\vec{r}' \cdot \vec{M}(\vec{r}')) - \frac{\partial(r'^2 \vec{\nabla} \cdot \vec{M}(\vec{r}'))}{r' \partial r'} \right]. \quad (2.38)$$

When applying integration by parts, the $\vec{\nabla}^2$ is replaced by $-k^2$ and by Green's theorem one can cast the radial derivative onto the spherical Bessel function. This results in

$$a_{lm}^{(M)} = \frac{-ik^2}{\sqrt{l(l+1)}} \sqrt{\frac{\mu_0}{\epsilon_0}} \int d\vec{r}' \left[j_l(kr') Y_{lm}^*(\theta', \phi') \left(\vec{\nabla} \cdot (\vec{r}' \times \vec{J}(\vec{r}')) - k^2 (\vec{r}' \cdot \vec{M}(\vec{r}')) \right) + Y_{lm}^*(\theta', \phi') \vec{\nabla} \cdot \vec{M}(\vec{r}') \frac{\partial(r' j_l(kr'))}{\partial r'} \right]. \quad (2.39)$$

As the source dimensions are small compared to the wavelength ($kr \leq 1$), the argument of the Bessel function is small. In this limit, j_l can be approximated as

$$j_l(kr) \approx \frac{(kr)^l}{(2l+1)!!}. \quad (2.40)$$

Inserting this approximation in Eq. (2.39) and keeping only the lowest orders in kr (leading to the cancellation of the $k^2 (\vec{r}' \cdot \vec{M}(\vec{r}'))$ term), one obtains

$$a_{lm}^{(M)} = \frac{-ik^{l+2}}{(2l+1)!!} \sqrt{\frac{l+1}{l}} \sqrt{\frac{\mu_0}{\epsilon_0}} (M_{lm} + M'_{lm}), \quad (2.41)$$

with the magnetic multipole moments defined as

$$M_{lm} = \frac{1}{l+1} \int d\vec{r}' r'^l Y_{lm}^*(\theta', \phi') \vec{\nabla} \cdot (\vec{r}' \times \vec{J}(\vec{r}')), \text{ and} \quad (2.42)$$

$$M'_{lm} = \int d\vec{r}' r'^l Y_{lm}^*(\theta', \phi') \vec{\nabla} \cdot \vec{M}(\vec{r}').$$

M_{lm} corresponds to the effective magnetization, while M'_{lm} accounts for the intrinsic magnetization $\vec{M}(\vec{r})$.

A similar procedure can be carried out in order to determine the electric multipole coefficients $a_{lm}^{(E)}$ (Ref. [5]). The result is

$$a_{lm}^{(E)} = \frac{-ick^{l+2}}{(2l+1)!!} \sqrt{\frac{l+1}{l}} [Q_{lm} + Q'_{lm}], \quad (2.43)$$

with the electric multipole moments

$$\begin{aligned} Q_{lm} &= \int d\vec{r}' r'^l Y_{lm}^*(\theta', \phi') \rho(\vec{r}') \\ Q'_{lm} &= \frac{-ik}{c(l+1)} \int d\vec{r}' r'^l Y_{lm}^*(\theta', \phi') \vec{\nabla} \cdot (\vec{r}' \times \vec{M}(\vec{r}')). \end{aligned} \quad (2.44)$$

The Poynting vector $\vec{S}$ is used to infer the angular dependent energy emitted per second and unit area. It is defined as

$$\vec{S}(\vec{r}, t) = \vec{E}(\vec{r}, t) \times \vec{H}(\vec{r}, t). \quad (2.45)$$

In the far field one has $|\vec{H}| = |\vec{B}|/\mu_0 = \sqrt{\epsilon_0/\mu_0} |\vec{E}|$ and the Poynting vector transforms to

$$|\vec{S}(\vec{r}, t)| = \sqrt{\frac{\epsilon_0}{\mu_0}} |\vec{E}(\vec{r}, t)|^2. \quad (2.46)$$

Applying the definition of the electric field (following Ref. [6]) as

$$\vec{E}(\vec{r}, t) = \vec{E}(\vec{r})e^{-i\omega t} + \vec{E}^*(\vec{r})e^{i\omega t}, \quad (2.47)$$

the time average takes the value

$$\langle |\vec{E}(\vec{r}, t)|^2 \rangle = 2 \cdot |\vec{E}(\vec{r})|^2, \quad (2.48)$$

and thus

$$\langle |\vec{S}(\vec{r}, t)| \rangle = 2 \sqrt{\frac{\epsilon_0}{\mu_0}} |\vec{E}(\vec{r})|^2. \quad (2.49)$$

From Eq. (2.35) we obtain for a single magnetic multipole of order lm

$$\vec{E}_{lm}^{(M)}(\vec{r}) = a_{lm}^{(M)} h_l^{(1)}(kr) \vec{X}_{lm}(\theta, \phi), \quad (2.50)$$

with $h_l^{(1)}$ the spherical Hankel functions and $\vec{X}_{lm}(\theta, \phi)$ the vector spherical harmonics. For far distances from the source ($kr \leq l$), the spherical Hankel functions can be approximated as

$$h_l^{(1)} \approx \frac{e^{i(kr-l\pi/2)}}{kr}. \quad (2.51)$$

Thus the total amount of energy radiated per second is calculated to be

$$\begin{aligned}
 P^{(M)} &= \int_{\Omega} < |\vec{S}(\vec{r}, t)| > d\Omega \\
 &= 2\sqrt{\frac{\epsilon_0}{\mu_0}} \int_{\Omega} \frac{|a_{lm}^{(M)}|^2}{k^2} |\vec{X}_{lm}(\theta, \phi)|^2 d\Omega \\
 &= 2\sqrt{\frac{\epsilon_0}{\mu_0}} \frac{|a_{lm}^{(M)}|^2}{k^2},
 \end{aligned} \tag{2.52}$$

where in the last step the normalization condition of the vector spherical harmonics was used. Inserting Eq. (2.41) for $a_{lm}^{(M)}$ and dividing by $\hbar\omega$, the rate of emission of a photon of frequency ω per unit time is derived.

$$\begin{aligned}
 A^{(M)} &= \frac{P^{(M)}}{\hbar\omega} = \frac{P^{(M)}}{\hbar ck} \\
 &= \frac{2\epsilon_0}{\hbar k^3} \cdot |a_{lm}^{(M)}|^2 \\
 &= \frac{2\mu_0}{\hbar} \frac{k^{2l+1}}{[(2l+1)!!]^2} \frac{l+1}{l} B_{eg}(Ml),
 \end{aligned} \tag{2.53}$$

where $B_{eg}(Ml)$ denotes the nuclear transition matrix element of multipole order l , defined for the transition from the excited state e to the ground state g as

$$B_{eg}(Ml) = (M_{lm} + M'_{lm})^2. \tag{2.54}$$

This matrix element can only be determined experimentally, however, Weisskopf derived a rough estimate for $B_{eg}(Ml)$, based on the single-particle nuclear shell model, leading to [6]

$$B_{eg}(Ml) \approx \frac{10}{\pi} \left(\frac{3}{3+l} \right)^2 R^{2l-2} \mu_N^2. \tag{2.55}$$

Here $R = 1.2 \cdot A_N^{1/3}$ fm is the nuclear radius (with A_N the mass number) and $\mu_N = 5.051 \cdot 10^{-27}$ J/T denotes the nuclear magneton. Equation (2.53) is often given in gaussian units, where it has to be multiplied by $4\pi/\mu_0$. The results of Eq. (2.55) can easily deviate by two orders of magnitude from the experimental results. In order to account for this, a correction factor $B_{W.u.}$ is introduced, corresponding to the transition rate in Weisskopf units. There is no complete agreement for $B_{W.u.}$ in case of ^{229m}Th in literature and the proposed values vary by a factor of 10 between $0.33 \cdot 10^{-2}$ and $4.55 \cdot 10^{-2}$ [7].

Similar to Eq. (2.53), the photon emission probabilities can also be estimated for electric multipole moments. In this case one obtains

$$\begin{aligned}
A^{(E)} &= \frac{2\mu_0}{\hbar k^3} \cdot |a_{lm}^{(E)}|^2 \\
&= \frac{2}{\hbar \epsilon_0} \frac{k^{2l+1}}{[(2l+1)!!]^2} \frac{l+1}{l} B_{eg}(El),
\end{aligned} \tag{2.56}$$

where $B_{eg}(El) = (Q_{lm} + Q'_{lm})^2$ and can be approximated as [6]

$$B_{eg}(El) \approx \frac{1}{4\pi} \left(\frac{3}{3+l} \right)^2 R^{2l} e^2. \tag{2.57}$$

During the radiative transition from an initial nuclear state i to a final state f , the overall angular momentum is conserved. This leads to the selection rules for multipole radiation l, m during nuclear transitions as

$$\begin{aligned}
|j_i - j_f| &\leq l \leq j_i + j_f \\
m_i - m_f &= m.
\end{aligned} \tag{2.58}$$

A further selection rule originates from parity conservation. Denoting the parity of the initial and final nuclear states as π_i and π_f , respectively, one obtains

$$\begin{aligned}
\pi_i &= \pi_f & \text{for } \pi = +1, \\
\pi_i &= -\pi_f & \text{for } \pi = -1,
\end{aligned} \tag{2.59}$$

where π denotes the parity of the multipole radiation. From the definition of electric and magnetic multipole radiation (see e.g. Ref. [6]) the parity π is obtained as

$$\begin{aligned}
\pi &= (-1)^l & \text{for electric multipoles,} \\
\pi &= -(-1)^l & \text{for magnetic multipoles.}
\end{aligned} \tag{2.60}$$

In the specific case of $^{229\text{m}}\text{Th}$, the nuclear transition occurs between the Nilsson states $3/2^+631$ (excited state) to $5/2^+633$ (ground state). Correspondingly, one obtains $|3/2 - 5/2| \leq l \leq 3/2 + 5/2$ and l can take values between 1 and 4. As decays are dominated by the lowest multipole order, only the multipole order $l = 1$ will be considered. From parity conservation it is seen that $\pi = +1$, which can, under the assumption $l = 1$, only be fulfilled for magnetic multipole radiation. Thus the transition of $^{229\text{m}}\text{Th}$ to the ground state will be mostly of type $M1$, with a small admixture of $E2$. The photon transition rate is calculated based on Eq. (2.53) to be $A \approx 4.9 \cdot 10^{-5} \text{ s}^{-1}$, conservatively assuming $B_{\text{W.u.}} = 0.33 \cdot 10^{-2}$. As the purely radiative lifetime τ_γ relates to the photon transition rate via

$$\tau_\gamma = \frac{1}{A}, \tag{2.61}$$

this corresponds to a lifetime of $\tau_\gamma \approx 2.0 \cdot 10^4 \text{ s}$, which is 5.6 h.

2.2.2 Internal Conversion

Besides γ decay of an excited nuclear state, internal conversion (IC) can occur, which is for most nuclear transitions the only relevant competing decay channel. During the internal conversion process, the nuclear excitation energy is transferred to an atomic shell electron, which is subsequently ejected from the atomic shell. Due to energy conservation, the electron's kinetic energy after the IC process (E_e) corresponds to the energy of the nuclear transition E_γ minus the binding energy of the electron E_b : $E_e = E_\gamma - E_b$. Thus internal conversion can only occur if the electron's binding energy is below the nuclear excitation energy. This is of particular importance for the case of $^{229\text{m}}\text{Th}$ with an extremely low nuclear excitation energy of about 7.8 eV. In this case it is expected that already for singly charged thorium ions the internal conversion process is energetically forbidden, as the ionization energy of Th^{1+} is 11.9 eV and therefore above the isomeric energy. Only for neutral thorium atoms, with an ionization energy of 6.3 eV, IC is expected to be the dominant decay channel.

It has to be pointed out that, during the IC process, the nucleus couples directly to the atomic shell by means of the exchange of a virtual photon. Internal conversion is thus a second order process, representing a competing decay channel to the first order process of the direct photonic decay (see also Fig. 3.5). In this way, the existence of an internal conversion decay channel leads to a lifetime reduction of the excited nuclear state. Opposed to that, it is not correct to imagine internal conversion as the emission of a (real) photon by nuclear deexcitation followed by a photo effect in the atomic shell. In the latter case the lifetime of the nuclear excited state would not be affected [6].

A complete theoretical derivation of the internal conversion process is involved and beyond the scope of this thesis. The interested reader is referred to the original work of Tralli and Goertzel [8]. Some problems concerning the gauge invariance were later resolved by G. Kramer [9]. For a description of the IC process we follow closely the lines of Rose (Refs. [10, 11]).

As nuclear γ decay and internal conversion are competing decay channels of an excited nuclear state, an internal conversion coefficient α_{ic} is defined as the decay rate via internal conversion (A_{ic}) divided by the γ -decay rate (A_γ). It can be shown [8, 9], that the IC coefficient is calculated (in SI units¹) as

$$\alpha_{\text{ic}} = \frac{A_{\text{ic}}}{A_\gamma} = \pi^2 k \lambda_c \alpha \cdot S \left| \int_0^\infty \Psi_f^* \left(\vec{\alpha} \cdot \vec{B}_{lm}(kr) + \phi_{lm}(kr) \right) \Psi_i r^2 dr d\Omega \right|^2, \quad (2.62)$$

where $k = E_\gamma/\hbar c$ is the wavenumber corresponding to the nuclear transition energy, $\lambda_c = 2\pi\hbar/m_e c \approx 2.426 \cdot 10^{-12}$ m denotes the Compton wavelength and $\alpha = e^2/4\pi\epsilon_0\hbar c \approx 1/137$ is the finestructure constant. S is an abbreviation for the sum over all unobserved degrees of freedom of the radiation, an average over initial and

¹Natural units are often used in literature, setting $\hbar = c = m_e = 1$. In this case $\lambda_c = 2\pi$, $\alpha = e^2$ and k and e are dimensionless.

a summation over final magnetic substates of the electron and Ψ_i and Ψ_f represent the electronic wave functions of the initial and final state that will be introduced below. $\vec{\alpha}$ corresponds to the Dirac matrix vector and $\vec{B}_{lm}$ and ϕ_{lm} are the vector and scalar potentials of an outgoing electromagnetic wave, respectively. They take different forms for electric and magnetic radiation of multipole order l . For magnetic radiation one has

$$\begin{aligned}\vec{B}_{lm}^{(M)} &= \sqrt{\frac{2}{\pi}} h_l^{(1)}(kr) \vec{X}_{lm}(\theta, \phi), \\ \phi_{lm}^{(M)} &= 0.\end{aligned}\tag{2.63}$$

Here $h_l^{(1)}$ denote the spherical Hankel function, and $\vec{X}_{lm}$ the vector spherical harmonics. Similarly, for electric multipole radiation, one has

$$\begin{aligned}\vec{B}_{lm}^{(E)} &= \sqrt{\frac{2}{\pi} \frac{1}{l(l+1)}} h_{l-1}^{(1)}(kr) \left(r \vec{\nabla} + \frac{lr}{r} \right) Y_{lm}(\theta, \phi), \\ \phi_{lm}^{(E)} &= i \sqrt{\frac{2}{\pi} \frac{l}{l+1}} h_l^{(1)}(kr) Y_{lm}(\theta, \phi),\end{aligned}\tag{2.64}$$

where Y_{lm} are the scalar spherical harmonics.

In Eq. (2.62), ψ_i and ψ_f represent relativistic electron wave functions of the central field, defined by the quantum numbers L , $J = L \pm 1/2$ and the magnetic quantum number μ . These are solutions of the Dirac equation for the Coulomb potential and can be separated into radial and angular components [10]

$$\psi = \begin{pmatrix} -i f_{LJ}(r) \Omega_{J\mu}^{(\pm)}(\theta, \phi) \\ g_{LJ}(r) \Omega_{J\mu}^{(\mp)}(\theta, \phi) \end{pmatrix} \quad \text{for } J = L \pm 1/2.\tag{2.65}$$

Here $\Omega_{J\mu}^{(\pm)}(\theta, \phi)$ correspond to the spin spherical harmonics, representing two-component spinors defined as

$$\Omega_{J\mu}^{(\pm)}(\theta, \phi) = \begin{pmatrix} C_{J\pm 1/2, \mu-1/2, 1/2, 1/2}^{J\mu} Y_{J\pm 1/2, \mu-1/2}(\theta, \phi) \\ C_{J\pm 1/2, \mu+1/2, 1/2, -1/2}^{J\mu} Y_{J\pm 1/2, \mu+1/2}(\theta, \phi) \end{pmatrix},\tag{2.66}$$

where C denotes the corresponding Clebsch–Gordan coefficient and Y_{lm} are the scalar spherical harmonics. The radial spinor wave functions $f(r)$ and $g(r)$ for the Coulomb potential can be calculated explicitly and are given in Ref. [12]. For the integral in Eq. (2.62), one obtains in case of magnetic multipole moments

$$\begin{aligned}
K^{(M)} &= \int_0^\infty \Psi_f^* \left(\vec{\alpha} \cdot \vec{B}_{lm}(kr) + \phi_{lm}(kr) \right) \Psi_i r^2 dr d\Omega \\
&= \frac{1}{\sqrt{l(l+1)}} \sqrt{\frac{2}{\pi}} \int_0^\infty \Psi_f^* h_l^{(1)}(kr) \vec{\alpha} \vec{L} Y_{lm}(\theta, \phi) \Psi_i r^2 dr d\Omega,
\end{aligned} \tag{2.67}$$

where the definition of the vector spherical harmonics as $\vec{X}_{lm}(\theta, \phi) = 1/\sqrt{l(l+1)} \vec{L} Y_{lm}(\theta, \phi)$ was used. Inserting $\vec{\alpha}$ as

$$\vec{\alpha} = \begin{pmatrix} 0 & \vec{\sigma} \\ \vec{\sigma} & 0 \end{pmatrix} \tag{2.68}$$

and using Eq. (2.65) for Ψ_i and Ψ_f one obtains

$$K^{(M)} = \frac{i}{\sqrt{l(l+1)}} \sqrt{\frac{2}{\pi}} \left[R_1 \int_\Omega \Omega_{J\mu}^{*(\pm)} \vec{\sigma} \vec{L} Y_{lm} \Omega_{J'\mu'}^{(\mp)} d\Omega - R_2 \int_\Omega \Omega_{J\mu}^{*(\mp)} \vec{\sigma} \vec{L} Y_{lm} \Omega_{J'\mu'}^{(\pm)} d\Omega \right]. \tag{2.69}$$

Here the radial integrals R_1 and R_2 were defined as

$$\begin{aligned}
R_1 &= \int_0^\infty f_{LJ}(r) h_l^{(1)}(kr) g_{L'J'}(r) r^2 dr \\
R_2 &= \int_0^\infty g_{LJ}(r) h_l^{(1)}(kr) f_{L'J'}(r) r^2 dr.
\end{aligned} \tag{2.70}$$

These integrals can only be calculated numerically. Using the identity [10]

$$\vec{\sigma} \cdot \left(\vec{L} Y_{lm} \right) \Omega_{J'\mu'} = \vec{\sigma} \cdot \vec{L} (Y_{lm} \Omega_{J'\mu'}) - Y_{lm} \vec{\sigma} \cdot \vec{L} \Omega_{J'\mu'}, \tag{2.71}$$

the integrals in Eq. (2.69) can be simplified in the following way

$$\begin{aligned}
\int_\Omega \Omega_{J\mu}^{*(\pm)} \vec{\sigma} \vec{L} Y_{lm} \Omega_{J'\mu'}^{(\mp)} d\Omega &= \int_\Omega \vec{\sigma} \vec{L} \Omega_{J\mu}^{*(\pm)} Y_{lm} \Omega_{J'\mu'}^{(\mp)} d\Omega - \int_\Omega \Omega_{J\mu}^{*(\pm)} Y_{lm} \vec{\sigma} \vec{L} \Omega_{J'\mu'}^{(\mp)} d\Omega \\
&= (\kappa + \kappa') \int_\Omega \Omega_{J\mu}^{*(\pm)} Y_{lm} \Omega_{J'\mu'}^{(\mp)} d\Omega.
\end{aligned} \tag{2.72}$$

In the last step the equations

$$\begin{aligned}
(\vec{\sigma} \vec{L} + 1) \Omega_{J\mu}^{*(\pm)} &= (L - J)(2J + 1) \Omega_{J\mu}^{*(\pm)} \quad \text{for } J = L \pm 1/2 \\
(\vec{\sigma} \vec{L} + 1) \Omega_{J'\mu'}^{(\mp)} &= -(L' - J')(2J' + 1) \Omega_{J'\mu'}^{(\mp)} \quad \text{for } J' = L' \pm 1/2
\end{aligned} \tag{2.73}$$

were used and κ (and κ') were introduced as

$$\begin{aligned}\kappa &= (L - J)(2J + 1) & \text{for } J = L \pm 1/2, \\ \kappa' &= (L' - J')(2J' + 1) & \text{for } J' = L' \pm 1/2.\end{aligned}\quad (2.74)$$

Similarly, for the second term in Eq. (2.69) one obtains

$$\begin{aligned}\int_{\Omega} \Omega_{J\mu}^{*(\mp)} \vec{\sigma} \bar{L} Y_{lm} \Omega_{J'\mu'}^{(\pm)} d\Omega &= -(\kappa + \kappa') \int_{\Omega} \Omega_{J\mu}^{*(\mp)} Y_{lm} \Omega_{J'\mu'}^{(\pm)} d\Omega \\ &= -(\kappa + \kappa') \int_{\Omega} \Omega_{J\mu}^{*(\pm)} Y_{lm} \Omega_{J'\mu'}^{(\mp)} d\Omega.\end{aligned}\quad (2.75)$$

Here, for the last step the square of a Pauli matrix $\sigma^2 = 1$ was introduced and the hermitian property of σ in combination with $\sigma \Omega_{J\mu}^{(\pm)} = -\Omega_{J\mu}^{(\mp)}$ was used [10]. Inserting the results of Eqs. (2.72) and (2.75) into Eq. (2.69) and using that Y_{lm} is a scalar function, the result is

$$K^{(M)} = \frac{i}{\sqrt{l(l+1)}} \sqrt{\frac{2}{\pi}} (R_1 + R_2) (\kappa + \kappa') \int_{\Omega} Y_{lm} \Omega_{J\mu}^{*(\pm)} \Omega_{J'\mu'}^{(\mp)} d\Omega. \quad (2.76)$$

From the definition of $\Omega_{J\mu}$ it is seen that

$$\Omega_{J\mu}^{*(\pm)} \Omega_{J'\mu'}^{(\mp)} = \sum_{\tau=\pm 1/2} C_{J\pm 1/2 \mu-\tau}^{J\mu} C_{J'\mp 1/2 \mu'-\tau}^{J'\mu'} Y_{J\pm 1/2 \mu-\tau}^* Y_{J'\mp 1/2 \mu'-\tau}. \quad (2.77)$$

For the following, we use that $L' = J' \mp 1/2$ and define $\bar{L} = J \pm 1/2$. The last equation contains the product of two spherical harmonics, which can be expressed in terms of a single spherical harmonic as [10]

$$Y_{\bar{L}\mu-\tau}^* Y_{L'\mu'-\tau} = \sum_{\nu} (-1)^{\mu-\tau} \sqrt{\frac{(2\bar{L}+1)(2L'+1)}{4\pi(2\nu+1)}} C_{\bar{L}0 L'0}^{\nu 0} C_{\bar{L}-\mu+\tau L' \mu'-\tau}^{\nu \mu'-\mu} Y_{\nu \mu'-\mu}. \quad (2.78)$$

Here the relation $Y_{Lm}^* = (-1)^m Y_{L-m}$ was applied. Inserting this into Eq. (2.77) leads to

$$\Omega_{J\mu}^{*(\pm)} \Omega_{J'\mu'}^{(\mp)} = \sum_{\nu} \sqrt{\frac{(2\bar{L}+1)(2L'+1)}{4\pi(2\nu+1)}} C_{\bar{L}0 L'0}^{\nu 0} Y_{\nu \mu'-\mu} S_{\nu}, \quad (2.79)$$

where S_{ν} was defined as

$$S_{\nu} = \sum_{\tau} (-1)^{\mu-\tau} C_{\bar{L} \mu-\tau 1/2 \tau}^{J\mu} C_{L' \mu'-\tau 1/2 \tau}^{J'\mu'} C_{\bar{L}-\mu+\tau L' \mu'-\tau}^{\nu \mu'-\mu}. \quad (2.80)$$

With some calculation, which will not be carried out at this point, it is possible to show that the sum over three Clebsch–Gordan coefficients can be explicitly expressed in terms of Racah coefficients W [10]

$$S_\nu = (-1)^{\bar{L}+L'+\nu+\mu+1/2} \sqrt{(2J+1)(2J'+1)} C_{J-\mu}^{\nu \mu'-\mu}{}_{J' \mu'} W(J \bar{L} J' L'; 1/2\nu). \quad (2.81)$$

Consequently, for the integral in Eq. (2.76) one obtains

$$\begin{aligned} \int_{\Omega} Y_{lm} \Omega_{J\mu}^{*(\pm)} \Omega_{J'\mu'}^{(\mp)} d\Omega &= \int_{\Omega} Y_{lm} \sum_{\nu} \sqrt{\frac{(2\bar{L}+1)(2L'+1)}{4\pi(2\nu+1)}} C_{\bar{L}0}^{\nu 0}{}_{L'0} Y_{\nu \mu'-\mu} S_{\nu} d\Omega \\ &= \sum_{\nu} \sqrt{\frac{(2\bar{L}+1)(2L'+1)}{4\pi(2\nu+1)}} C_{\bar{L}0}^{\nu 0}{}_{L'0} S_{\nu} (-1)^m \int_{\Omega} Y_{l-m}^* Y_{\nu \mu'-\mu} d\Omega \\ &= (-1)^m \sum_{\nu} \sqrt{\frac{(2\bar{L}+1)(2L'+1)}{4\pi(2\nu+1)}} C_{\bar{L}0}^{\nu 0}{}_{L'0} S_{\nu} \delta_{l,\nu} \delta_{m,\mu'-\mu} \\ &= (-1)^m \sqrt{\frac{(2\bar{L}+1)(2L'+1)}{4\pi(2l+1)}} C_{\bar{L}0}^{l 0}{}_{L'0} S_l \delta_{\mu,\mu'+m}. \end{aligned} \quad (2.82)$$

Inserting Eq. (2.76) together with this integral into Eq. (2.62) the result is

$$\alpha_{ic}^{(M)} = \frac{k\lambda_c\alpha}{2} \cdot S \left| \frac{(R_1 + R_2)(\kappa + \kappa')}{\sqrt{l(l+1)}} \sqrt{\frac{(2\bar{L}+1)(2L'+1)}{(2l+1)}} C_{\bar{L}0}^{l 0}{}_{L'0} S_l \delta_{\mu,\mu'+m} \right|^2. \quad (2.83)$$

The sum S , which represents the sum over all final states and the average over all initial states, can be explicitly written as $S = 1/(2l+1) \sum_{\mu\mu'mJ}$ [10], leading to

$$\begin{aligned} \alpha_{ic}^{(M)} &= \frac{k\lambda_c\alpha}{2} \frac{1}{2l+1} \sum_{\mu\mu'mJ} \left| \frac{(R_1 + R_2)(\kappa + \kappa')}{\sqrt{l(l+1)}} \sqrt{\frac{(2\bar{L}+1)(2L'+1)}{(2l+1)}} C_{\bar{L}0}^{l 0}{}_{L'0} S_l \delta_{\mu,\mu'+m} \right|^2 \\ &= \frac{k\lambda_c\alpha}{2} \frac{2L'+1}{l(l+1)(2l+1)^2} \sum_J |R_1 + R_2|^2 (\kappa + \kappa')^2 (2\bar{L}+1) |C_{\bar{L}0}^{l 0}{}_{L'0}|^2 \sum_{\mu\mu'} S_l^2 \\ &= \frac{k\lambda_c\alpha}{2} \frac{(2L'+1)(2J'+1)}{l(l+1)(2l+1)^2} \sum_J |R_1 + R_2|^2 (\kappa + \kappa')^2 (2\bar{L}+1)(2J+1) \\ &\quad |C_{\bar{L}0}^{l 0}{}_{L'0}|^2 W^2(J \bar{L} J' L'; 1/2l) \sum_{\mu\mu'} |C_{J-\mu}^{\mu'-\mu}{}_{J' \mu'}|^2. \end{aligned} \quad (2.84)$$

Applying now the relation

$$\sum_{\mu\mu'} |C_{J-\mu}^{\mu'-\mu}{}_{J' \mu'}|^2 = 2l+1 \quad (2.85)$$

leads to the final result for the internal conversion coefficient as

$$\alpha_{ic}^{(M)} = \frac{k\lambda_c\alpha}{2} \frac{(2L' + 1)(2J' + 1)}{l(l + 1)(2l + 1)} \sum_J |R_1 + R_2|^2 (\kappa + \kappa')^2 (2\bar{L} + 1)(2J + 1) |C_{\bar{L}0 \ L'0}^{l \ 0}|^2 W^2(J\bar{L}J'L'; 1/2l). \quad (2.86)$$

Let us now consider the special case of $^{229\text{m}}\text{Th}$: Corresponding calculations were for the first time performed by Strizhov and Tkalya [13] and more recently also by Karpeshin and Trzhaskovskaya [14]. Due to the expected energy value of 7.8 eV, internal conversion is expected to occur only in neutral ^{229}Th and only by coupling to the two outermost electrons, populating the $6d_{3/2}$ and the $7s_{1/2}$ electronic levels (usually IC electrons are generated in the innermost shells). Calculation reveals, however, that the internal conversion process is dominated by the $7s_{1/2}$ shell [13]. Thus one can assume $L' = 0$ and $J' = 1/2$. As the nuclear transition is of type $M1$, we have $l = 1$ and Eq. (2.86) simplifies to

$$\alpha_{ic} = \frac{k\lambda_c\alpha}{6} \sum_J |R_1 + R_2|^2 (\kappa + \kappa')^2 (2\bar{L} + 1)(2J + 1) |C_{\bar{L}0 \ 1/2 \ 0}^{1 \ 0}|^2 W^2(J\bar{L}1/2 \ 0; 1/2 \ 1). \quad (2.87)$$

This equation provides the basis for numerical calculations resulting in $\alpha_{ic} \approx 10^9$ [13, 14]. Some minor dependency on the applied atomic shell model was found [14]. Assuming a purely photonic lifetime $\tau_\gamma = 2.0 \cdot 10^4 \text{ s}$ (see Sect. 2.2.1), the expected lifetime of $^{229\text{m}}\text{Th}$ under internal conversion in the neutral thorium atom is $\tau_{ic} \approx 20 \mu\text{s}$. This value is in good agreement with the recently measured lifetime $\tau_{ic} \approx 10 \mu\text{s}$ (corresponding to $t_{1/2} \approx 7 \mu\text{s}$) [15].

2.3 Nuclear Laser Excitation

One of the main motivations for studying $^{229\text{m}}\text{Th}$ is that it is the only known nuclear state that allows for direct nuclear laser excitation using already existing laser technology. In the following, the theory of nuclear laser excitation is discussed. In the first section the optical Bloch equations are derived. In the second section an expression for the number of excited nuclei in saturation is obtained, assuming the low-saturation limit. This equation is applied to the special case of $^{229\text{m}}\text{Th}$ in the third section. In the last section, the case of $^{235\text{m}}\text{U}$, which exhibits the second lowest excitation energy of 76 eV, is discussed for comparison.

2.3.1 The Optical Bloch Equations

Quite generally the optical Bloch equations describe the coupling of coherent light to a two-level system [16]. For an atomic two-level system usually electric dipole transitions are discussed. For nuclear two-level systems, however, higher multipolarities play a more pronounced role and have to be taken into consideration. The optical Bloch equations for a nuclear two-level system consisting of degenerate ground and excited nuclear states will be derived in the following. Starting point for this derivation is the quantum optical master equation in Lindblad form given in the Schrödinger picture as [17, 18]

$$\frac{\partial \hat{\rho}}{\partial t} = -\frac{i}{\hbar} [\hat{H}, \hat{\rho}] + \mathcal{L}[\hat{\rho}], \quad (2.88)$$

where the Lindblad super-operator $\mathcal{L}[\hat{\rho}]$ is defined as

$$\mathcal{L}[\hat{\rho}] = \sum_{m_g m_e} \Gamma_{m_g m_e} \left(\hat{A}_{m_g m_e} \hat{\rho} \hat{A}_{m_g m_e}^\dagger - \frac{1}{2} \hat{\rho} \hat{A}_{m_g m_e}^\dagger \hat{A}_{m_g m_e} - \frac{1}{2} \hat{A}_{m_g m_e}^\dagger \hat{A}_{m_g m_e} \hat{\rho} \right). \quad (2.89)$$

The first term on the right-hand side of Eq. (2.88) corresponds to the von-Neumann equation, which describes the time propagation of the density operator $\hat{\rho}$ without energy dissipation. The Lindblad super-operator, models the decay of the excited states due to coupling to the environment. The summation is performed over all possible deexcitations from the nuclear excited to the nuclear ground states, taking into consideration the magnetic sublevels. For simplicity we have neglected the decoherences between different sublevels of the ground and excited state. The total angular momentum quantum numbers of the ground and excited states are denoted by j_g and j_e , respectively, with the corresponding magnetic quantum numbers m_g and m_e satisfying $-j_g \leq m_g \leq j_g$ and $-j_e \leq m_e \leq j_e$. Furthermore, $\Gamma_{m_g m_e}$ is the partial decay rate from the excited nuclear sublevel described by the magnetic quantum number m_e to the ground nuclear sublevel described by the magnetic quantum number m_g . The Lindblad operators $\hat{A}_{m_g m_e}$ are the annihilation operators of the excited states, explicitly given as

$$\hat{A}_{m_g m_e} = e^{-iE_g t/\hbar} |j_g, m_g\rangle \langle j_e, m_e| e^{+iE_e t/\hbar}. \quad (2.90)$$

It is assumed that the total Hamiltonian of the system $\hat{H}$ can be expressed as $\hat{H} = \hat{H}_N + \hat{H}_I$, with $\hat{H}_N$ the nuclear Hamiltonian and $\hat{H}_I$ the resonant light-interaction Hamiltonian, which describes the interaction of laser light of angular frequency ω_L with a nuclear transition of the same angular frequency $\omega = \omega_L$.

The Hilbert space vectors of the ground and excited states $|j_g, m_g\rangle$ and $|j_e, m_e\rangle$ are eigenstates to the nuclear Hamiltonian without interaction term $\hat{H}_N |j_i, m_i\rangle = E_i |j_i, m_i\rangle$ with $i \in \{g, e\}$, where E_g and E_e denote the energy of the ground and

excited nuclear state. We therefore have $(E_e - E_g)/\hbar = \omega$. The time dependency of the states can be gained back by multiplying with $e^{-iE_g t/\hbar}$ or $e^{-iE_e t/\hbar}$, respectively.

In the Schrödinger picture and for the considered case, the density operator $\hat{\rho}$ takes the explicit form

$$\begin{aligned}\hat{\rho}(t) = & \sum_{m_e} \rho_{ee}(m_e) e^{-iE_e t/\hbar} |j_e, m_e\rangle \langle j_e, m_e| e^{+iE_e t/\hbar} \\ & + \sum_{m_g m_e} \rho_{ge}(m_g, m_e) e^{-iE_g t/\hbar} |j_g, m_g\rangle \langle j_e, m_e| e^{+iE_e t/\hbar} \\ & + \sum_{m_g m_e} \rho_{eg}(m_e, m_g) e^{-iE_e t/\hbar} |j_e, m_e\rangle \langle j_g, m_g| e^{+iE_g t/\hbar} \\ & + \sum_{m_g} \rho_{gg}(m_g) e^{-iE_g t/\hbar} |j_g, m_g\rangle \langle j_g, m_g| e^{+iE_g t/\hbar}.\end{aligned}\quad (2.91)$$

Here $\rho_{ee}(m_e)$ and $\rho_{gg}(m_g)$ denote the population probabilities of the ground and excited nuclear sublevels, respectively and $\rho_{ge}(m_g, m_e) = \rho_{eg}^*(m_g, m_e)$ are the so called coherences. From this it is inferred that the population probability of the excited sublevel characterized by the magnetic quantum number $\tilde{m}_e$ is

$$\rho_{ee}(m_e) = \langle j_e, m_e | e^{+iE_e t/\hbar} \hat{\rho} e^{-iE_e t/\hbar} | j_e, m_e \rangle. \quad (2.92)$$

For an explicit calculation we will first consider the von-Neumann-term of Eq. (2.88). This transforms to

$$-\frac{i}{\hbar} [\hat{H}, \hat{\rho}] = -\frac{i}{\hbar} [\hat{H}_N + \hat{H}_I, \hat{\rho}] \quad (2.93)$$

Calculating the matrix element $\rho_{ee}(m_e)$ of the density operator by making use of Eq. (2.92) and inserting the unity operator $\hat{\mathbb{1}}$ between each operator product of Eq. (2.93) as

$$\begin{aligned}\hat{\mathbb{1}} = & \sum_{m_g} e^{-iE_g t/\hbar} |j_g, m_g\rangle \langle j_g, m_g| e^{+iE_g t/\hbar} \\ & + \sum_{m_e} e^{-iE_e t/\hbar} |j_e, m_e\rangle \langle j_e, m_e| e^{+iE_e t/\hbar},\end{aligned}\quad (2.94)$$

the von-Neumann-term gives

$$\begin{aligned}& -\frac{i}{\hbar} \langle j_e, m_e | e^{+iE_e t/\hbar} [\hat{H}, \hat{\rho}] e^{-iE_e t/\hbar} | j_e, m_e \rangle \\ = & -\frac{i}{\hbar} \sum_{m_g} \left[\rho_{ge}(m_g, m_e) e^{i\omega t} \langle j_e, m_e | \hat{H}_I | j_g, m_g \rangle \right. \\ & \left. - \rho_{eg}(m_g, m_e) e^{-i\omega t} \langle j_g, m_g | \hat{H}_I | j_e, m_e \rangle \right].\end{aligned}\quad (2.95)$$

Here $\langle j_e, m_e | \hat{H}_I | j_e, m_e \rangle = 0$ was used, as $\hat{H}_I$ describes only transitions between the nuclear ground and excited sublevels.

In a next step, the extra term in Eq. (2.88) as imposed by the Lindblad super-operator is considered. Using Eq. (2.92) for the matrix element $\rho_{ee}(m_e)$ and inserting Eq. (2.90) for $\hat{A}_{m_g m_e}$, a direct calculation reveals that this term is of the form

$$\begin{aligned} & -\frac{i}{\hbar} \langle j_e, m_e | e^{+iE_e t/\hbar} \mathcal{L}[\hat{\rho}] e^{-iE_e t/\hbar} | j_e, m_e \rangle \\ & = -\rho_{ee}(m_e) \sum_{m_g} \Gamma_{m_g m_e}. \end{aligned} \quad (2.96)$$

Performing the same calculation also for the remaining matrix elements $\rho_{gg}(m_g)$ and $\rho_{ge}(m_g, m_e) = \rho_{eg}^*(m_g, m_e)$ leads to the optical Bloch equations for degenerate ground and excited states including damping [19]² (see also Refs. [20, 21])

$$\begin{aligned} \dot{\rho}_{ee}(m_e) &= -\frac{i}{\hbar} \sum_{m_g} \left[\rho_{ge}(m_g, m_e) e^{i\omega t} \langle j_e, m_e | \hat{H}_I | j_g, m_g \rangle - \right. \\ & \quad \left. \rho_{eg}(m_g, m_e) e^{-i\omega t} \langle j_g, m_g | \hat{H}_I | j_e, m_e \rangle \right] - \rho_{ee}(m_e) \sum_{m_g} \Gamma_{m_g m_e}, \\ \dot{\rho}_{ge}(m_g, m_e) &= \frac{i}{\hbar} \left[\rho_{gg}(m_g) - \rho_{ee}(m_e) \right] e^{-i\omega t} \langle j_g, m_g | \hat{H}_I | j_e, m_e \rangle - \rho_{ge}(m_g, m_e) \left(i\Delta + \sum_{\tilde{m}_g} \tilde{\Gamma}_{\tilde{m}_g m_e} \right), \\ \dot{\rho}_{gg}(m_g) &= \frac{i}{\hbar} \sum_{m_e} \left[\rho_{ge}(m_g, m_e) e^{i\omega t} \langle j_e, m_e | \hat{H}_I | j_g, m_g \rangle - \right. \\ & \quad \left. \rho_{eg}(m_g, m_e) e^{-i\omega t} \langle j_g, m_g | \hat{H}_I | j_e, m_e \rangle \right] + \sum_{m_e} \rho_{ee}(m_e) \Gamma_{m_g m_e}. \end{aligned} \quad (2.97)$$

In these equations the possibility of detuning of the laser with respect to the transition frequency is taken into account by introducing $\Delta = \omega - \omega_L$, where ω_L denotes the angular frequency of the laser light. The decay rate of the coherences was introduced as $\tilde{\Gamma}_{m_g m_e} = \Gamma_{m_g m_e}/2$.

For a specific multipolarity l , the partial decay rates $\Gamma_{m_g m_e}$ can be explicitly expressed as [1, 19]

$$\Gamma_{m_g m_e} = \sum_{\sigma} \frac{2j_e + 1}{2l + 1} |C_{j_g m_g j_e - m_e}^{l\sigma}|^2 \Gamma_{\text{tot}}, \quad (2.98)$$

where $C_{j_g m_g j_e - m_e}^{l\sigma}$ is the corresponding Clebsch–Gordan coefficient, σ denotes the photon angular momentum projection and Γ_{tot} is the total decay rate of the excited

²Note, that the sum over $\tilde{m}_g$ is missing in the second equation in Ref. [19].

state, including both radiative decay and internal conversion channels. Using this definition, summation over the final states m_g together with the average over the initial states m_e for the partial decay rates renders the total decay rate Γ_{tot} . From Eq. (2.98) and using the summation properties of the Clebsch–Gordan coefficients, one obtains

$$\begin{aligned} \sum_{m_g} \Gamma_{m_g m_e} &= \sum_{m_g, \sigma} \frac{2j_e + 1}{2l + 1} |C_{j_g m_g j_e - m_e}^{l\sigma}|^2 \Gamma_{\text{tot}} \\ &= \Gamma_{\text{tot}}. \end{aligned} \quad (2.99)$$

Consequently, also the sum over the decay rates of the coherences can be simplified to

$$\begin{aligned} \sum_{m_g} \tilde{\Gamma}_{m_g m_e} &= \frac{1}{2} \sum_{m_g} \Gamma_{m_g m_e} = \frac{1}{2} \Gamma_{\text{tot}} \\ &= \tilde{\Gamma}_{\text{tot}}. \end{aligned} \quad (2.100)$$

Here the total decay rate of the coherences $\tilde{\Gamma}_{\text{tot}}$ was introduced. A short coherence time of the laser pulse introduces an additional decay rate of the coherence terms in Eqs. (2.97) [22, 23], such that $\tilde{\Gamma}_{\text{tot}} = (\Gamma_{\text{tot}} + \Gamma_L)/2$ where Γ_L denotes the bandwidth of the laser light [21].

In its general form, the optical Bloch equations given in Eq. (2.97) can only be solved numerically. Under certain conditions, however, an analytical solution is possible. The analytical solution for a realistic scenario of nuclear laser excitation will be discussed in the following section.

2.3.2 The Low-Saturation Limit

In this section the optical Bloch equations given in Eq. (2.97) will be analytically solved under the assumption that the nuclear transition is driven with moderate laser intensities and a laser bandwidth broader than the natural linewidth of the nuclear transition. In this case the low-saturation limit ($\sum_{m_g} \rho_{gg}(m_g) \approx 1$ and $\rho_{gg}(m_g) \gg \rho_{ee}(m_e)$) is valid and the initial population of the ground-state magnetic sublevels can be approximated as

$$\rho_{gg}(m_g) \approx \frac{1}{2j_g + 1}. \quad (2.101)$$

Further, for a bandwidth of the laser light significantly broader than the linewidth of the nuclear transition ($\Gamma_L \gg \Gamma_{\text{tot}}$) one obtains $\tilde{\Gamma}_{\text{tot}} \approx \Gamma_L/2$ and the coherences ρ_{ge} will relax fast to an equilibrium with $\dot{\rho}_{ge}(m_g, m_e) = 0$. Under these assumptions the equation for $\dot{\rho}_{ge}$ of Eq. (2.97) simplifies to

$$0 = \frac{i}{\hbar} \frac{e^{-i\omega t}}{2j_g + 1} \langle j_g, m_g | \hat{H}_I | j_e, m_e \rangle - \rho_{ge}(m_g, m_e) (i\Delta + \tilde{\Gamma}_{\text{tot}}), \quad (2.102)$$

which is known as the adiabatic elimination method [24], and one obtains

$$\rho_{ge}(m_g, m_e) = \frac{i}{\hbar} \frac{e^{-i\omega t}}{2j_g + 1} \frac{\langle j_g, m_g | \hat{H}_I | j_e, m_e \rangle}{i\Delta + \tilde{\Gamma}_{\text{tot}}}. \quad (2.103)$$

Inserting this expression into the equation for $\dot{\rho}_{ee}$ leads to

$$\begin{aligned} \dot{\rho}_{ee}(m_e) &= \frac{1}{\hbar^2(2j_g + 1)} \sum_{m_g} \left[\frac{|\langle j_g, m_g | \hat{H}_I | j_e, m_e \rangle|^2}{i\Delta + \tilde{\Gamma}_{\text{tot}}} + \frac{|\langle j_g, m_g | \hat{H}_I | j_e, m_e \rangle|^2}{-i\Delta + \tilde{\Gamma}_{\text{tot}}} \right] - \rho_{ee}(m_e) \Gamma_{\text{tot}} \\ &= \frac{2}{\hbar^2(2j_g + 1)} \frac{\tilde{\Gamma}_{\text{tot}}}{\Delta^2 + \tilde{\Gamma}_{\text{tot}}^2} \sum_{m_g} |\langle j_g, m_g | \hat{H}_I | j_e, m_e \rangle|^2 - \rho_{ee}(m_e) \Gamma_{\text{tot}}. \end{aligned} \quad (2.104)$$

In the rotating wave approximation H_I can be regarded as time independent. Therefore this differential equation is solved for $\rho_{ee}(t = 0) = 0$ by

$$\rho_{ee}(m_e) = \frac{2\tilde{\Gamma}_{\text{tot}} [1 - e^{-\Gamma_{\text{tot}} \cdot t}]}{\hbar^2 \Gamma_{\text{tot}} (2j_g + 1) (\Delta^2 + \tilde{\Gamma}_{\text{tot}}^2)} \sum_{m_g} |\langle j_g, m_g | \hat{H}_I | j_e, m_e \rangle|^2. \quad (2.105)$$

Consequently, from Eq. (2.105) the excited state population density is obtained by summation over m_e and reads

$$\begin{aligned} \rho_{ee} &= \sum_{m_e} \rho_{ee}(m_e) \\ &= \frac{2\tilde{\Gamma}_{\text{tot}} [1 - e^{-\Gamma_{\text{tot}} \cdot t}]}{\hbar^2 \Gamma_{\text{tot}} (2j_g + 1) (\Delta^2 + \tilde{\Gamma}_{\text{tot}}^2)} \sum_{m_g m_e} |\langle j_g, m_g | \hat{H}_I | j_e, m_e \rangle|^2. \end{aligned} \quad (2.106)$$

This excited state population corresponds to a small (arbitrary) frequency fraction $\Delta\omega \leq \Gamma_{\text{tot}}$ of the total frequency width of the laser light Γ_L . In order to calculate the full population density, ρ_{ee} has to be divided by $\Delta\omega$ and integrated over the full laser linewidth Γ_L . Only the overlap with the transition linewidth will contribute to this integral. For this reason, the integration limits can be extended to infinity without changing the value of the integral. Thus we have

$$\begin{aligned}
\rho_{ee}^{\text{tot}} &= \int \frac{\rho_{ee}}{\Delta\omega} d\omega_L \\
&= \int d\omega_L \frac{2\tilde{\Gamma}_{\text{tot}} [1 - e^{-\Gamma_{\text{tot}} \cdot t}]}{\hbar^2 \Gamma_{\text{tot}} \Delta\omega (2j_g + 1) (\Delta^2 + \tilde{\Gamma}_{\text{tot}}^2)} \sum_{m_g m_e} |\langle j_g, m_g | \hat{H}_I | j_e, m_e \rangle|^2 \\
&= \frac{2[1 - e^{-\Gamma_{\text{tot}} \cdot t}]}{\hbar^2 \Gamma_{\text{tot}} \Delta\omega (2j_g + 1)} \sum_{m_g m_e} |\langle j_g, m_g | \hat{H}_I | j_e, m_e \rangle|^2 \int d\omega_L \frac{\tilde{\Gamma}_{\text{tot}}}{\Delta^2 + \tilde{\Gamma}_{\text{tot}}^2} \\
&= \frac{2\pi [1 - e^{-\Gamma_{\text{tot}} \cdot t}]}{\hbar^2 \Gamma_{\text{tot}} \Delta\omega (2j_g + 1)} \sum_{m_g m_e} |\langle j_g, m_g | \hat{H}_I | j_e, m_e \rangle|^2,
\end{aligned} \tag{2.107}$$

where in the last step the integral

$$\frac{1}{\pi} \int d\omega_L \frac{\tilde{\Gamma}_{\text{tot}}}{\Delta^2 + \tilde{\Gamma}_{\text{tot}}^2} = 1 \tag{2.108}$$

was applied.

The interaction matrix element $\langle j_g, m_g | \hat{H}_I | j_e, m_e \rangle$ can be expressed in terms of the reduced interaction matrix element $\langle j_g \| \hat{H}_I \| j_e \rangle$ by means of the Wigner–Eckart theorem [25]

$$\langle j_g, m_g | \hat{H}_I | j_e, m_e \rangle = C_{j_e m_e l \sigma}^{j_g m_g} \langle j_g \| \hat{H}_I \| j_e \rangle. \tag{2.109}$$

From the summation properties of the Clebsch–Gordan coefficients one has [26]

$$\sum_{m_g, m_e} |C_{j_e m_e l \sigma}^{j_g m_g}|^2 = \frac{2j_g + 1}{2l + 1} \tag{2.110}$$

and therefore

$$\begin{aligned}
\sum_{m_g, m_e} |\langle j_g, m_g | \hat{H}_I | j_e, m_e \rangle|^2 &= \sum_{m_g, m_e} |C_{j_e m_e l \sigma}^{j_g m_g}|^2 |\langle j_g \| \hat{H}_I \| j_e \rangle|^2 \\
&= \frac{2j_g + 1}{2l + 1} |\langle j_g \| \hat{H}_I \| j_e \rangle|^2.
\end{aligned} \tag{2.111}$$

Defining the Rabi frequency Ω as

$$\Omega = \frac{2|\langle j_e \| \hat{H}_I \| j_g \rangle|}{\sqrt{2l + 1}\hbar} = \sqrt{\frac{2j_g + 1}{2j_e + 1}} \frac{2|\langle j_g \| \hat{H}_I \| j_e \rangle|}{\sqrt{2l + 1}\hbar} \tag{2.112}$$

and inserting Eq.(2.111) into Eq.(2.107), the resulting expression for ρ_{ee}^{tot} reads

$$\rho_{ee}^{\text{tot}} = \frac{\pi \Omega^2}{2\Gamma_{\text{tot}} \Delta\omega} \frac{2j_e + 1}{2j_g + 1}. \tag{2.113}$$

A detailed description for the derivation of the explicit form of the interaction matrix element $\langle j_g, m_g | \hat{H}_I | j_e, m_e \rangle$ is given in Refs. [1, 27] and will not be carried out at this point. The result for magnetic multipole radiation is [19]³

$$|\langle j_g, m_g | \hat{H}_I | j_e, m_e \rangle| = \frac{E_0 \sqrt{2\pi}}{c} \sqrt{\frac{(2l+1)(l+1)}{l}} \frac{k^{l-1}}{(2l+1)!!} \sqrt{\frac{2j_e+1}{2j_g+1}} |C_{j_e m_e l \sigma}^{j_g m_g}| \sqrt{B_{eg}(ML)}, \quad (2.115)$$

similarly, for electric multipole radiation, we have

$$|\langle j_g, m_g | \hat{H}_I | j_e, m_e \rangle| = E_0 \sqrt{2\pi} \sqrt{\frac{(2l+1)(l+1)}{l}} \frac{k^{l-1}}{(2l+1)!!} \sqrt{\frac{2j_e+1}{2j_g+1}} |C_{j_e m_e l \sigma}^{j_g m_g}| \sqrt{B_{eg}(El)}. \quad (2.116)$$

Here E_0 is the amplitude of the electric driving field and approximations for the transition matrix elements $B_{eg}(ML)$ and $B_{eg}(El)$ were given in Eqs. (2.55) and (2.57), respectively.

In the following, only the magnetic multipole term will be discussed. By comparison with Eq. (2.109), it is clear that the reduced interaction Hamiltonian is obtained from Eq. (2.115) by dropping the Clebsch–Gordan coefficient. According to Eq. (2.112), the Rabi frequency is thus inferred to be

$$\Omega = \frac{\sqrt{8\pi} E_0}{\hbar c} \sqrt{\frac{(l+1)}{l}} \frac{k^{l-1}}{(2l+1)!!} \sqrt{B_{eg}(ML)}. \quad (2.117)$$

The transition matrix element $B_{eg}(ML)$ can be related to the transition rate $A^{(M)}$ by means of Eq. (2.53) as

$$B_{eg}(ML) = \frac{\hbar}{2\mu_0} \frac{l[(2l+1)!!]^2}{l+1} \left(\frac{1}{k}\right)^{2l+1} A^{(M)}. \quad (2.118)$$

³Note, that compared to Ref. [19] a further factor of $\sqrt{2l+1/2j_g+1}$ occurs. This difference originates from a different Clebsch–Gordan coefficient. Using the notation of Ref. [19], redefining $-\sigma$ to σ and interchanging the ground and excited state, one obtains

$$\begin{aligned} |C(j_g j_e l; m_g - m_e \sigma)| &= |C_{j_g m_g j_e - m_e}^{l \sigma}| = \sqrt{2l+1} \left| \begin{pmatrix} j_g & j_e & l \\ m_g & -m_e & -\sigma \end{pmatrix} \right| = \sqrt{2l+1} \left| \begin{pmatrix} j_g & j_e & l \\ -m_g & m_e & \sigma \end{pmatrix} \right| \\ &= \sqrt{2l+1} \left| \begin{pmatrix} j_e & l & j_g \\ m_e & \sigma & -m_g \end{pmatrix} \right| = \frac{\sqrt{2l+1}}{\sqrt{2j_g+1}} |C_{j_e m_e l \sigma}^{j_g m_g}|. \end{aligned} \quad (2.114)$$

Here some relations for the Wigner-3j symbols were used [26]. Only if the Clebsch–Gordan coefficient is in the final form, the Wigner–Eckart theorem (2.109) can be applied to define the reduced interaction Hamiltonian.

For the derivation of Eq. (2.115) the electric field was assumed to be of the form [27]

$$E(x, t) = E_0 \left(e^{i(k_0 x - \omega_0 t)} + e^{-i(k_0 x - \omega_0 t)} \right) = 2E_0 \cos(k_0 x - \omega_0 t). \quad (2.119)$$

The time-averaged energy per unit volume of the electromagnetic wave is consequently given as

$$\begin{aligned} \rho &= \frac{\epsilon_0 \overline{|E(x, t)|^2}}{2} + \frac{\overline{|B(x, t)|^2}}{2\mu_0} \\ &= \epsilon_0 \overline{|E(x, t)|^2} = 2\epsilon_0 E_0^2, \end{aligned} \quad (2.120)$$

where $B(x, t) = E(x, t)/c$ and $\overline{\cos^2(k_0 x - \omega_0 t)} = 1/2$ was used. In order to obtain E_0 from the spectral energy density per frequency interval ρ^ω , one has to divide ρ by the arbitrary but small frequency fraction $\Delta\omega \leq \Gamma_{\text{tot}}$.

$$\rho^\omega = \frac{\rho}{\Delta\omega}. \quad (2.121)$$

Correspondingly E_0 can be expressed as

$$E_0 = \sqrt{\frac{\rho^\omega \Delta\omega}{2\epsilon_0}}. \quad (2.122)$$

Using $k = \omega/c$, where ω denotes the frequency of the nuclear transition and inserting Eqs. (2.122) and (2.118) in Eq. (2.117), one obtains for the Rabi frequency

$$\Omega = \sqrt{\frac{2\pi c^3 \rho^\omega \Delta\omega A^{(M)}}{\hbar\omega^3}}. \quad (2.123)$$

This expression is inserted in Eq. (2.113) in order to infer the total number of excited nuclei in saturation for a given spectral energy density ρ^ω of the laser light

$$\begin{aligned} N_{\text{sat}} &= \rho_{ee}^{\text{tot}} \cdot N_0 \\ &= \frac{\pi\Omega^2 N_0}{2\Gamma_{\text{tot}}\Delta\omega} \frac{2j_e + 1}{2j_g + 1} \\ &= \frac{\rho^\omega \pi^2 c^3 A^{(M)} N_0}{\Gamma_{\text{tot}} \hbar\omega^3} \frac{2j_e + 1}{2j_g + 1}. \end{aligned} \quad (2.124)$$

Here N_0 denotes the total number of irradiated nuclei. Also the time dependency can be regained. From Eq. (2.105) one obtains

$$N(t) = N_{\text{sat}} (1 - e^{-\Gamma_{\text{tot}} t}). \quad (2.125)$$

The same result also holds for electric multipole transitions, however, with $A^{(M)}$ interchanged by $A^{(E)}$. We therefore write for the radiative transition rate

$$A^{(M)} = A^{(E)} = A = \frac{1}{\tau_\gamma}, \quad (2.126)$$

where τ_γ denotes the radiative lifetime of the excited nuclear state. Opposed to that, Γ_{tot} corresponds to the total natural decay rate of the transition, which also includes potential non-radiative decay branches. In case of a potential internal conversion decay channel of the excited state, this means

$$\Gamma_{\text{tot}} = (1 + \alpha_{\text{ic}})A, \quad (2.127)$$

where α_{ic} denotes the internal conversion coefficient.

It should be pointed out that the same result Eq. (2.125) can also be obtained via the Einstein rate equations, which are valid for the considered case of the low-saturation limit and a transition linewidth smaller than the width of the laser light. The Einstein rate equations under resonant irradiation read [28]

$$\begin{aligned} \dot{N}_g &= (1 + \alpha_{\text{ic}})AN_e + \rho^\omega B^\omega N_e - \rho^\omega B^\omega \frac{g_e}{g_g} N_g \\ \dot{N}_e &= -(1 + \alpha_{\text{ic}})AN_e - \rho^\omega B^\omega N_e + \rho^\omega B^\omega \frac{g_e}{g_g} N_g, \end{aligned} \quad (2.128)$$

where N_g and N_e are the population numbers of the ground and excited state, respectively, A and B^ω denote the Einstein coefficients corresponding to spontaneous emission (A) as well as stimulated emission and absorption (B^ω) [28, 29]. $g_g = 2j_g + 1$ and $g_e = 2j_e + 1$ are the degeneracies of the ground and excited state and ρ^ω denotes the spectral energy density of the electromagnetic field which drives the transition. Solving this equation leads to the number of excited nuclei given as

$$N_e(t) = \frac{\rho^\omega B^\omega \frac{g_e}{g_g} N_0}{(1 + \alpha_{\text{ic}})A + (1 + \frac{g_e}{g_g})\rho^\omega B^\omega} \left(1 - e^{-[(1 + \alpha_{\text{ic}})A + (1 + \frac{g_e}{g_g})\rho^\omega B^\omega]t} \right), \quad (2.129)$$

The Einstein B^ω coefficient is related to A via [28]

$$B^\omega = \frac{\pi^2 c^3}{\hbar \omega^3} \cdot A. \quad (2.130)$$

In case that $B^\omega \rho^\omega \ll A$ is fulfilled, Eq. (2.129) transforms to

$$\begin{aligned}
N_e(t) &= \frac{\rho^\omega B^\omega \frac{g_e}{g_g} N_0}{(1 + \alpha_{ic}) A} (1 - e^{-(1 + \alpha_{ic}) A t}) \\
&= \frac{\rho^\omega \pi^2 c^3 A N_0}{\Gamma_{\text{tot}} \hbar \omega^3} \frac{2j_e + 1}{2j_g + 1} (1 - e^{-\Gamma_{\text{tot}} t}),
\end{aligned} \tag{2.131}$$

where in the last step Eq. (2.130) was inserted for B^ω and $\Gamma_{\text{tot}} = (1 + \alpha_{ic}) A$ was used. This result is identical with Eq. (2.125), derived via the optical Bloch equations, and will be applied to $^{229\text{m}}\text{Th}$ in the following.

2.3.3 Laser Excitation of $^{229\text{m}}\text{Th}$

In the previous section it was shown that, during laser irradiation, the number of excited nuclei in saturation N_{sat} can be calculated via

$$N_{\text{sat}} = \frac{\rho^\omega \pi^2 c^3 A N_0}{\Gamma_{\text{tot}} \hbar \omega^3} \frac{2j_e + 1}{2j_g + 1} \tag{2.132}$$

where ρ^ω denotes the spectral energy density of the laser light, $A = 1/\tau_\gamma$ is the radiative transition rate, $\Gamma_{\text{tot}} = (1 + \alpha_{ic})A$ the total transition rate with α_{ic} as the internal conversion coefficient and ω the angular frequency of the nuclear transition. It is concluded that N_{sat} is independent of the lifetime of the excited state. However, the time after which saturation is reached is lifetime dependent. From Eq. (2.131) the time dependence is known to be of the form

$$N_e(t) = N_{\text{sat}} (1 - e^{-\Gamma_{\text{tot}} t}). \tag{2.133}$$

Thus the characteristic saturation time τ_{sat} is

$$\tau_{\text{sat}} = \frac{1}{\Gamma_{\text{tot}}} = \frac{1}{(1 + \alpha_{ic})A} = \frac{\tau_\gamma}{1 + \alpha_{ic}}. \tag{2.134}$$

Based on Eq. (2.133) the nuclear excitation rate can be approximated as

$$R \approx \dot{N}_e(t)|_{t=0} = \Gamma_{\text{tot}} N_{\text{sat}} = \frac{\rho^\omega \pi^2 c^3 A N_0}{\hbar \omega^3} \frac{2j_e + 1}{2j_g + 1}. \tag{2.135}$$

The spectral energy density ρ^ω can be related to the laser intensity I of the laser light via

$$\rho^\omega = \frac{I}{c \Gamma_L}, \tag{2.136}$$

with Γ_L the bandwidth of the laser light. In the nuclear clock concept it is envisaged to excite a single ion ($N_0 = 1$) in a Paul trap. The laser excitation should be carried

out with a single mode of a frequency comb with a bandwidth of approximately $\Gamma_L = 2\pi$ Hz and the nuclear excitation rate should be of about the same value $R \approx \Gamma_L$. Thus the condition for the required laser intensity for the development of a single-ion nuclear clock reads

$$I \approx \frac{\hbar\omega^3\Gamma_L^2}{\pi^2c^2A} \frac{2j_g + 1}{2j_e + 1}. \quad (2.137)$$

Inserting the values of $A = 10^{-4}$ Hz [30], $\omega = 1.19 \cdot 10^{16}$ Hz, $j_g = 5/2$ and $j_e = 3/2$ for ^{229}Th results in $I = 0.012$ W/cm². Assuming that the single mode of the frequency comb could be focused onto an area of $10 \mu\text{m}^2$, the required mode power for the development of a nuclear clock based on ^{229}Th would be $\rho \approx 12$ nW. This is a realistic power for a single mode of a frequency comb.

A similar result can also be obtained via the Rabi frequency Ω . From the theory of Rabi oscillations it is known, that these can occur under the condition that $\tilde{\Gamma}_{\text{tot}}/2 < \Omega$ [31]. Here $\tilde{\Gamma}_{\text{tot}} = (\Gamma_{\text{tot}} + \Gamma_L)/2$ denotes the total decay rate of the coherences, with Γ_{tot} the linewidth of the transition and Γ_L the bandwidth of the laser light. From Eq. (2.123) in combination with Eq. (2.121) and $\rho = I/c$, the Rabi frequency is obtained as

$$\Omega = \sqrt{\frac{2\pi c^2 I A}{\hbar\omega^3}}. \quad (2.138)$$

Accordingly, the condition for the observation of Rabi oscillations can be written as

$$\sqrt{\frac{2\pi c^2 I A}{\hbar\omega^3}} > \frac{\max(\Gamma_{\text{tot}}, \Gamma_L)}{4}. \quad (2.139)$$

In the case of ^{229}Th ions in a Paul trap the internal conversion process is energetically suppressed and thus $\alpha_{\text{ic}} = 0$ holds (see Sect. 2.2.2). Therefore we have $\Gamma_L \gg \Gamma_{\text{tot}}$ and the above condition is securely fulfilled for $\Omega = \Gamma_L$. This leads to a required laser intensity in order to drive Rabi oscillations of

$$I \approx \frac{\hbar\omega^3\Gamma_L^2}{2\pi c^2 A}. \quad (2.140)$$

For ^{229}Th we obtain the same result as above of $I = 0.012$ W/cm². Changes to these considerations could originate from the fact that, even if internal conversion is suppressed, electronic bridge effects might still be present (see Sect. 3.5.1) and have to be considered by a further electronic bridge coefficient α_{eb} that cannot be set to zero. Values of α_{eb} between 10 (Th^{3+}) and 10^5 (Th^{1+} in case of resonance) have to be expected, depending on the charge state and transition energy under consideration [32–34]. These effects will, however, only play a role if the nuclear transition linewidth would be broadened beyond the bandwidth of the laser light used for nuclear excitation. Further, the electric field strength at the point of the nucleus

may not equal the field strength of the incoming laser wave, as (anti-)shielding effects from the atomic shell will play a role (e.g. in case of Th^{4+} a magnetic dipole anti-shielding factor of 217.3 was found [35]).

From the above discussions it is clear that nuclear laser excitation of $^{229\text{m}}\text{Th}$ appears possible, especially as there are strong ongoing developments of VUV-laser technology. The generation of coherent VUV light providing an average output power of 0.5 mW at 149 nm via intra-cavity 7th harmonic generation in noble gas of the 1040 nm light produced by an Yb-fiber laser system was recently reported [36]. Considering that $\sim 10^5$ comb modes are generated in this way, the estimated average mode power is 5 nW, sufficiently large to drive the ^{229}Th nuclear transition when focused to an area of about $10 \mu\text{m}^2$.

The discussed laser excitation scheme requires a precise knowledge of the isomer's transition energy and does not allow for a search of the resonance frequency, given the large uncertainty in transition energy of about 1 eV. This has led to the general assumption that the direct nuclear laser excitation of $^{229\text{m}}\text{Th}$ requires a better knowledge of the isomer's excitation energy. In the following, a new laser excitation scheme for $^{229\text{m}}\text{Th}$ is presented that allows to circumvent this requirement, in this way providing the basis for a first nuclear laser excitation of $^{229\text{m}}\text{Th}$. The presented method is making use of the short-lived internal conversion decay channel, thereby allowing for nearly background-free detection of electrons emitted in the isomeric decay [37].

A tunable and pulsed VUV laser system with an energy of $E_L \approx 10 \mu\text{J}$ per pulse, $R_L = 10 \text{ Hz}$ repetition rate, 5 ns pulse duration at a beam area of $A_L = 1 \text{ mm}^2$ and a linewidth of $\Delta\nu_L = 10 \text{ GHz}$ as is already available [38] is assumed for irradiation of a thin (2.5 nm) layer of ^{229}Th atoms on a surface of 1 mm^2 . Under this condition, the isomeric decay will occur by internal conversion with a half-life of $7 \mu\text{s}$, resulting in an IC decay factor of $\alpha_{\text{ic}} \approx 10^9$ [15]. As the spectral energy density of a single laser pulse is significantly larger than the time-averaged laser energy density and also the number of irradiated thorium atoms is large ($N_0 \approx 7.6 \cdot 10^{13}$), a significant amount of nuclei can be expected to be excited during a single laser pulse. For the spectral energy density of the laser pulse one obtains

$$\rho^\omega = \frac{E_L}{c T_L A_L \cdot \Delta\omega_L} \approx 1.1 \cdot 10^{-10} \text{ Js/m}^3. \quad (2.141)$$

Here $T_L = 5 \text{ ns}$ denotes the pulse duration, the remaining laser parameters were taken as stated above. As the laser linewidth of 10 GHz is still significantly larger than the IC-broadened nuclear transition linewidth of about 15.9 kHz, Eq. (2.132) for the number of excited nuclei in saturation is valid and one obtains

$$N_{\text{sat}} = \frac{2}{3} \frac{\rho^\omega \pi^2 c^3 N_0}{(1 + \alpha_{\text{ic}}) \hbar \omega^3} \approx 4.2 \cdot 10^3. \quad (2.142)$$

As these nuclear excitations will decay with a characteristic half-life of about $7 \mu\text{s}$ after the laser pulse via internal conversion, it is possible to trigger the detector

in accordance with the laser pulses in order to allow for a nearly background-free detection of the emitted electrons. These could be detected with an MCP detector, providing about 50% detection efficiency if the electrons are post-accelerated to about 300 eV kinetic energy. The only source of background that has to be considered are electrons, which are generated by the laser light on the surface. Electrons produced by the photo effect, however, appear on the timescale of nano-seconds and can therefore be distinguished from the comparatively long-lived isomeric decay. Due to the relatively low laser intensity, no thermal electron emission can be expected to occur.

In the described way, it should be possible to detect the isomeric decay by means of its characteristic lifetime within one single laser pulse and thus on a micro-second timescale. Even when assuming 100 laser pulses per scan step, the total time required for scanning the large energy range of 1 eV would only be $2.4 \cdot 10^5$ s, corresponding to 2.7 days. This detection scheme would pave the way for a first direct nuclear laser excitation of $^{229\text{m}}\text{Th}$ without the requirement of an improved isomeric energy value. The described detection technique can be considered as laser-driven conversion-electron Mössbauer spectroscopy (CEMS) [39] in the optical region. It could also provide the basis for a CEMS-based solid-state nuclear clock, however, with an IC-enlarged natural transition linewidth of expectedly about 15.9 kHz (corresponding to 7 μs half-life) [15].

2.3.4 A Comparison to $^{235\text{m}}\text{U}$

The second lowest nuclear excitation, which is known by today, is the isomeric state of ^{235}U with an excitation energy of about 76 eV [40]. This low excitation energy has led to the conclusion that also $^{235\text{m}}\text{U}$ could be a candidate for direct nuclear laser excitation, as soon as laser technology has evolved to deliver intense laser light at the required wavelength by high-harmonic generation [41, 42]. In this case, the long half-life of $^{235\text{m}}\text{U}$ of about 26 min would even make the isomer a good candidate for a nuclear clock.

Such considerations, however, do not take the multipolarity of the ^{235}U isomer-to-ground-state transition into account. Being a transition from the spin and parity values $1/2^+$ (excited state) to $7/2^-$ (ground state), the multipolarity is $E3$. According to the Weisskopf estimate Eqs. (2.56) and (2.57), the radiative transition rate $A^{(E)}$ is

$$\begin{aligned} A^{(E)} &\approx \frac{1}{2\pi\hbar\epsilon_0} \frac{k^{2l+1}}{[(2l+1)!!]^2} \frac{l+1}{l} \left(\frac{3}{3+l}\right)^2 R^{2l} e^2 \\ &= \frac{\omega^7 R^6 e^2}{66150\pi\hbar\epsilon_0 c^7} = 2.60 \cdot 10^{-23} \text{ s}^{-1}. \end{aligned} \quad (2.143)$$

This corresponds to a lifetime of $\tau_\gamma = 3.8 \cdot 10^{22}$ s or $1.2 \cdot 10^{15}$ years. The reason for the actually measured lifetime of $2.25 \cdot 10^3$ s is a huge internal conversion coefficient of estimatedly $\alpha_{\text{ic}} \approx 1.7 \cdot 10^{19}$. According to Eq. (2.140) the laser intensity required

to drive the transition with a single mode of a frequency comb $\Gamma_L = 2\pi$ Hz can be estimated to be

$$I \approx \frac{\hbar\omega^3\Gamma_L^2}{2\pi c^2 A} = 4.3 \cdot 10^{19} \text{ W/cm}^2. \quad (2.144)$$

Here the angular frequency $\omega = 1.15 \cdot 10^{17}$ Hz for $^{229\text{m}}\text{U}$ was used. This laser intensity is unrealistically large. The reason is, that only the radiative decay rate A enters the calculation and not the IC-broadened nuclear decay rate Γ_{tot} , which corresponds to the comparatively short half-life of 26 min.

The nuclear transition of third lowest energy is contained in ^{110}Ag and possesses an excitation energy of 1.1 keV ($t_{1/2} = 660$ ns) [43], which could be excited with the help of free-electron lasers, similar as the 14.4 keV Mössbauer transition of ^{57}Fe [44]. This technology, however, can by today not provide linewidths sufficiently small to allow for the development of nuclear frequency standards. For this reason, $^{229\text{m}}\text{Th}$ is currently the only known nucleus that would allow for the development of a nuclear clock applying already existing technology. This situation may change in the future, taking the fast development of laser technology into account [45–48].

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On the Direct Detection of 229m Th

Von der Wense, L.

2018, XIX, 224 p. 76 illus., 14 illus. in color., Hardcover

ISBN: 978-3-319-70460-9