Gain without inversion for gamma radiation

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Abstract

Gain without inversion for a level-mixing scheme is studied for gamma-optics. In this scheme nuclear level mixing is created by misalignment of a dc magnetic field with respect to the c-axis of a noncubic crystal. Axially symmetric electron nuclear coupling and nuclear quadrupole interaction with an electric field gradient produce two electro-nuclear levels that are equally mixed and split, with the energy gap dependent on the tilting angle of the magnetic field. By laser excitation of an electron transition, nuclear spin coherence can be created between these two levels. A condition is found for the predominant population of the dark state by spontaneous emission. This state is a particular superposition of the mixed states, such that, by selection rules, the transition from it cannot be excited by γ-radiation. If all absorbing nuclei are in the dark state, resonant γ-absorption is suppressed in the sample. At the same condition, γ-emission of the excited nuclei is allowed because the corresponding transition terminates in another component of the mixed states superposition. The constraints on the tilting angle and reciprocal gap between two mixed electro-nuclear levels are found, setting the limits to these values beyond which the gain without inversion becomes impossible. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

Among the basic problems of the gamma-laser implementation one can find the smallness of the nuclear cross sections, a small photon flux and a small concentration of the excited nuclei at available pump powers. Moreover, an estimation of the pump power necessary to achieve population inversion required for gamma-lasing shows that the sample can even explode at this power before lasing takes place.

One can find a review on recent proposals for gamma-ray lasers (grasers) in [1]. Since reaching and maintaining population inversion is not feasible with the available pumping schemes, alternative proposals of gamma-ray amplification with reduced pump requirements are necessary.

In the late 80’s lasing without inversion (LWI) has been proposed [2–4] for the optical domain. The key point for LWI is the significant reduction of the atomic absorption due to the quantum interference of transition paths. This interference leaves the emission process unchanged. Since absorption and emission become nonreciprocal, light amplification is possible even when the population of the upper level is less than that of the ground state.

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The first proposals on LWI were followed by a large number of theoretical publications on the subject (see, for example, the reviews [5–8]) where different schemes were studied. Experimental verification of inversionless amplification and lasing was successfully realized [9–16] and reports on new experiments related to this phenomenon continue to appear [17–20].

For gamma optics, gain without inversion (GWI) was considered in [21] where it was proposed to use the level mixing scheme in order to split the $\gamma$-transition in two paths. The same splitting occurs for the transition driven by the coherent pump. Transition splitting in several paths is well known in solid state spectroscopy as branching of the transitions [22]. It results in the modulation of the coherent signals such as spin echo and photon echo [22–24]. The modulation pattern contains information about the hyperfine structure of the electron states or resonant frequencies of nuclei present in the host crystal. The effect arises from terms of the form $S, I$, and $S, I$ in the Hamiltonian describing the interaction of the electron spins ($S$) with the nuclear spins ($I$) in their immediate neighborhood. Transition branching occurs due to the different symmetry of interactions included in the Hamiltonian. For the scheme proposed in [21] this branching becomes possible due to the level crossing (in the nuclear ground state) caused by the interplay between the Zeeman splitting (a magnetic dipole interaction) and the interaction between the nuclear quadrupole moment with an electric field gradient. The level crossing (in the nuclear excited state) with the successive mixing due to the small misalignment of a magnetic field with respect to an electric field gradient (known as anticrossing) was proposed in [25] for very accurate measurements of the hyperfine parameters. Because the dips in the gamma-quanta emission spectrum, seen along the crystal symmetry axis at the anticrossings, can be made very sharp, this effect was called level mixing resonance (LMR). LMR has been proved to be a powerful experimental method for the measurement of quadrupole moments of short lived isomers [26–28].

One can find a similarity between LMR, resulting in the spontaneous emission decrease or increase along a particular direction, and the amplification without inversion, since both effects involve the interference of quantum states. However, LMR takes place when the anticrossing occurs in the excited nuclear state, while GWI becomes possible when the anticrossings happen in the ground nuclear state. In the scheme of [21], the coherence of the mixed states was created by an RF pump. Unless particular relaxation mechanisms between nuclear hyperfine states could be invoked, driving by means of microwave radiation is not effective [29], because of nearly equal populations of the hyperfine sublevels, even at liquid helium temperature. It is also mentioned in review [1] that microwave radiation can only reduce but, unlike the case of the optical excitation for the optical lasing, cannot completely cancel the absorption. Since the solution of the graser dilemma requires many orders of magnitude reduction in pump power one has to turn to the consideration of the optical excitation creating a coherent superposition of a close pair of lower states.

Optical excitation of the electron-nuclear states opens new perspectives for gamma-GWI, because an optical pump is capable to create larger coherence of the nuclear hyperfine states [30]. Nuclear spin excitation by laser pumping, proposed in [30], is possible if level mixing takes place only in the ground state of the electron shell. The scheme of [30] is similar to the Raman driven laser without inversion, which was considered in [31,32]. Even though the physical processes in both schemes are similar, a discussion of the geometry of the intended experiment, symmetry constraints, relevant selection rules of the transitions involved as well as realistic values of their relaxation parameters is necessary. It should be mentioned that recently another electro-nuclear system, driven by a coherent optical field and probed at a gamma-transition, has been proposed [33]. In that scheme four- and five-level electro-nuclear systems are considered. The symmetry breaking between the pair of lower levels and the pair of upper levels is realized by hyperfine interactions. It should be also pointed out that the five-level scheme, proposed in [33], is similar to the scheme of [30], as it involves a coherent population trapping in the sublevels of the lowest level $|1\rangle$.

In this paper we present the analysis of the GWI conditions for the level mixing (LM) scheme in solids. In Section 2 our scheme with optical pump and gamma-probe is discussed. Section 3 presents
the master equations for the LM scheme and the analysis of the possibility of GWI for gamma-quanta in solids containing Mössbauer nuclei. The analytical solution of the master equations, its analysis and discussion of the influence of the ground state splitting on the GWI are presented in Section 4.

2. Level mixing scheme for gamma-optics

In general, the emission of a $\gamma$-quantum is followed by the recoil of the emitting nucleus, which spreads the emission line. This effect reduces the nuclear resonant cross section. In solids, some nuclei (so-called Mössbauer nuclei [34]) do not experience this effect as the whole crystalline lattice can absorb the recoil. Therefore, the Mössbauer nuclei, with their recoil-less gamma-ray transitions, are good candidates for a gamma-ray laser. Moreover, the concentration of the Mössbauer nuclei in the host crystal can be varied to a large extent. Following the proposition in [21], we take a noncubic uniaxial crystal as a host and a Mössbauer nucleus with a ground state spin higher than 1/2, e.g. $I_G = 3/2$, where the subscript $G$ refers to the nuclear ground state. We suppose that the excited nucleus has a different spin, for example, $I_E = 1/2$ ($E$ refers to the excited nuclear state). We study the gamma-quanta absorption or emission processes along the crystal symmetry axis $c$ if an external magnetic field $H_c$ parallel to this axis is applied. The interaction of the nuclear quadrupole moment with the electric field gradient of the host splits the four degenerate levels of the ground state nucleus into a degenerate doublet, i.e., $m = \pm 1/2$ and $m = \pm 3/2$. The Zeeman interaction with the field $H_z$ further splits each component of the doublet. Because the electric and magnetic interactions have different up-down symmetry, level crossings may occur. The energy diagram of the nucleus indicating the transitions we study is shown in Fig. 1(a).

If the propagation direction of the $\gamma$-quanta is parallel to $H_c$, only transitions $\Delta m = \pm 1$ are allowed, while the $\Delta m = 0$ transitions are forbidden. We choose the second level-crossing [in Fig. 1(a)] of the ground state nucleus as an initial state and the hyperfine sublevel $| \! \! - 1/2 \rangle$ of the excited nucleus as a final state of the $\gamma$-absorption transition. It should be pointed out that for the LM scheme, the spectral components of the $\gamma$-transition must be well resolved, i.e., in our case the transition from the second crossing to the state $| - 1/2 \rangle_E$ is to have no overlap with the $\gamma$-transition $| + 1/2 \rangle_G \rightarrow | - 1/2 \rangle_E$, where subscripts $G$ and $E$ are again introduced to distinguish hyperfine components in the ground ($G$) and the excited ($E$) nuclear states. Below by lower case letters $g$ and $e$ we denote the ground and excited states of the electron shell.

In the scheme proposed in [21] a small dc magnetic field $H_c \perp c$, risen by the misalignment of the magnetic field with respect to the $c$-axis, produces a breaking of the axial symmetry at and near the crossing field. It results in the mixing of the wave functions and a repulsion of the energy levels [see Fig. 1(b)]

$$|1\rangle = \cos \psi | - 1/2 \rangle_G - \sin \psi | - 3/2 \rangle_G ,$$

$$|2\rangle = \sin \psi | - 1/2 \rangle_G + \cos \psi | - 3/2 \rangle_G ,$$

where $\psi$ is the mixing angle ($\psi = \pi/4$ at exact crossing). One can find that both $\gamma$-transitions $|1\rangle \rightarrow | - 1/2 \rangle_E$ and $|2\rangle \rightarrow | - 1/2 \rangle_E$ become allowed in the $z$-direction ($z \parallel c$), while without mixing by the $H_c$-field, only the transition $| - 3/2 \rangle_G \rightarrow | - 1/2 \rangle_E$ is allowed and the other one, $| - 1/2 \rangle_G \rightarrow | - 1/2 \rangle_E$, is forbidden in this direction. In Fig. 1(b), the state $|4\rangle$ is chosen as the spin level $| - 1/2 \rangle_E$ of the excited nucleus and the transition path is split due to the level mixing. The lower case letter $g$ is introduced in the definition of the functions $|1\rangle$, $|2\rangle$ and $|4\rangle$ given in the Fig. 1(b) to indicate that the electron shell is not excited when the system is in these states. The details related to this aspect are discussed below.

The mixing of the levels and the effective change in selection rules for the $\gamma$-transition are caused by the fact that the transverse field $H_x$ is dominant at level crossing. Indeed, the effect of the field $H_x$ is compensated by the quadrupole interaction and $H_z$ produces the level mixing and repulsion. For the nuclear excited state the situation is completely different because, due to different hyperfine interaction, we are far from the crossing. For the case, shown in Fig. 1(a), this crossing takes place only at zero magnetic field.
In the next section it will be shown that optical pumping of the mixed states creates coherence between them. At certain conditions, spontaneous emission together with optical pumping lead to the trapping of the nucleus in a dark state \[|\rangle.\] In our case, the dark state corresponds to the symmetric combination of the spin states \(|1\rangle\) and \(|2\rangle\). The latter is a pure \(m\)-state \(|-1/2\rangle_G\), i.e.
\[
\cos \psi |1\rangle + \sin \psi |2\rangle = |-1/2\rangle_G.
\]
This state cannot be excited by \(\gamma\)-quanta parallel to the \(c\)-axis because of the selection rule. In this way the optical pump makes the anticrossing states non-absorbing.

In [30] it was proposed to pump the ground state nuclear spin and create coherence between the two mixed states by laser excitation of the electron shell coupled with the nuclear spin via hyperfine interaction. We assume axial symmetry of the hyperfine interaction, i.e., \(\mathcal{H}_{hyp} = A \sqrt{S \cdot I}\), where \(S\) and \(I\) are electron and nuclear spin operators, respectively, and \(A\) is an interaction constant. This constant as well as the electron spin are different in the ground and excited states of the electron shell. Therefore, the crossing conditions for the excited and ground states are not fulfilled at the same external magnetic field.

Because of the axial symmetry of the hyperfine interaction, the state function of the compound system can be factorized in a nuclear and an electronic wave function. The LM scheme is shown in Fig. 1(b). Capital letters \(G\) and \(E\) denote ground and excited nuclear states, whereas lowercase letters \(g\) and \(e\) are related to the ground and excited state of the electron shell, respectively. The longitudinal component of the external magnetic field, \(H_z\), to-

![Energy diagram of the nucleus interacting with γ-probe and laser-pump.](image-url)
gether with the z-component of the magnetic hyperfine field, \(H_y\), produced by the ground-state electron, create a level crossing of the ground-state nucleus, while the transverse component of the external field mixes and splits the levels into a doublet \(|G\rangle_{1}\) and \(|G\rangle_{2}\). When the electron is excited, the hyperfine field \((H_y)\) is different, so the spin levels \(-1/2\rangle_G\) and \(-3/2\rangle_G\) do not cross.

At the crossing point in the electron ground state, the main part of the Hamiltonian for the chosen states is defined by the interaction with a transverse component of the magnetic field \(H_y\). In the excited electron state, we are far from the crossing and the main interaction is determined by the interaction with the longitudinal component of the magnetic field (external plus hyperfine) and the quadrupole interaction. Therefore, the main parts of the Hamiltonians in the excited and ground electron states do not commute if anticrossing occurs only in one of the states. This is the main reason why laser excitation creates a superposition of the nuclear eigenstates \([24]\). The matrix element of the electric dipole operator \(d\) for the optical transition \(g \rightarrow e\) involves also a transition between different nuclear spin states

\[
d_{gm_e,em_g} = \langle g \mid \hat{\mu}_g \rangle \langle e \mid \hat{m}_g \rangle
\]

if the nuclear spin states \(|\hat{\mu}_g\rangle\) and \(|m_g\rangle\) related to the ground \(|g\rangle\) and excited \(|e\rangle\) electron states, respectively, are not orthogonal. Here we assume that the functions \(|\hat{\mu}_g\rangle\) and \(|m_g\rangle\) are defined so that relevant hyperfine Hamiltonians are diagonal. If these Hamiltonians do commute (or can be diagonalized in the same basis), then

\[
\langle \hat{\mu}_g \mid m_g \rangle = \delta_{\mu_g m_g},
\]

and the nuclear spin state is not changed by the optical excitation. Splitting or branching of the optical excitation path is an inherent feature of the LM scheme. Laser pumping along the c-axis of the crystal excites the electron transition to the state \(|G\rangle_{3}\) and, simultaneously, excites the coherence of the states \(|Gg\rangle_{1}\) and \(|Gg\rangle_{2}\) if the excitation frequency is resonant. Here \(|Gg\rangle_{1}\) and \(|Gg\rangle_{2}\) describe the ground state nucleus and ground state electron, \(|Eg\rangle_{4}\) is the excited state of the nucleus if the electron is not excited and \(|G\rangle_{3}\) is the state if the electron is excited and the nucleus is not. We do not consider the laser pumping of the excited nucleus electron shell as there is no level mixing of the nuclear spin in this state. The laser pumping, as has been explained, can create only a population redistribution among the spin levels of the excited nucleus, which can be incorporated into the incoherent pumping of the nucleus.

In the next sections we present the analysis of the solution of the master equations for the LM scheme along with the study of the GWI conditions at the level mixing.

3. Master equations for the level mixing scheme

Our LM-scheme is described by the density matrix \(\rho\) of the four-level system (see Fig. 1(b)) which satisfies the following master equations

\[
\dot{\rho}_{11} = i(C_{14}\sigma_{41} - C_{41}\sigma_{14}) + (B_{13}\sigma_{31} - B_{31}\sigma_{13}) - (W_{12} + P)\rho_{11} + \sum_{n \neq 1} W_{n1}\rho_{1n},
\]

\[
\dot{\rho}_{22} = i(C_{24}\sigma_{42} - C_{42}\sigma_{24}) + (B_{23}\sigma_{32} - B_{32}\sigma_{23}) - (W_{21} + P)\rho_{22} + \sum_{n \neq 2} W_{n2}\rho_{2n},
\]

\[
\dot{\rho}_{33} = -i(B_{13}\sigma_{31} - B_{31}\sigma_{13}) - i(B_{23}\sigma_{32} - B_{32}\sigma_{23}) - (W_{31} + W_{32})\rho_{33},
\]

\[
\dot{\rho}_{44} = -i(C_{14}\sigma_{41} - C_{41}\sigma_{14}) - i(C_{24}\sigma_{42} - C_{42}\sigma_{24}) - (W_{41} + W_{42})\rho_{44} + P(\rho_{11} + \rho_{22}),
\]

\[
\dot{\sigma}_{q4} = (i\Delta_q - \Gamma_q)\sigma_{q4} + iC_{q4}(\rho_{44} - \rho_{qq}) - iC_{q4}\rho_{q4} + iB_{q3}\sigma_{34},
\]

\[
\dot{\sigma}_{q3} = (i\Delta_q - \Gamma_q)\sigma_{q3} + iB_{q3}(\rho_{33} - \rho_{qq}) - iB_{q3}\rho_{q3} + iC_{q4}\sigma_{43},
\]

\[
\dot{\sigma}_{34} = [i(\Delta_q - \delta_q) - \Gamma_q]\sigma_{34} + i(B_{32}\sigma_{24} + B_{31}\sigma_{14}) - i(\sigma_{32}C_{24} + \sigma_{31}C_{14}),
\]

\[
\dot{\rho}_{12} = (i\omega_{24} - i\Gamma_{24})\rho_{12} + i(C_{14}\sigma_{42} - C_{42}\sigma_{14}) + i(B_{13}\sigma_{32} - B_{32}\sigma_{13}),
\]

where \(\rho_{ij}\) are the density matrix elements in the Schrödinger picture. Here states \(|1\rangle\) to \(|4\rangle\) are de-
fined in Fig. 1(b). The index \( \eta \) indicates the ground state \( 1 \) or \( 2 \). The nuclear and optical coherences are 

\[ \sigma_{\eta_1} = \rho_{\eta_1} \exp(-i\Omega t + iKz) \quad \text{and} \quad \sigma_{\eta_2} = \rho_{\eta_2} \exp(-i\omega t + iKz), \]

where \( (\Omega, K) \) and \( (\omega, K) \) are the frequencies and wave numbers of the \( \gamma \)-probe and laser pump fields, respectively. The \( z \)-axis coincides with the \( c \)-axis of the crystal, \( \Delta_0 = \omega_{\eta_1} - \Omega \) and \( \delta_0 = \omega_{\eta_2} - \omega \) are the detuning parameters of the probe and the pump with respect to the transition frequencies \( \omega_{\eta_1} \) (\( \gamma \)-transition) and \( \omega_{\eta_2} \) (optical transition), respectively. The rates \( \Gamma, \Gamma' \) and \( \Gamma_{3\eta} \) are the dephasing rates of the optical transition, \( \gamma \)-transition and ground state spin coherence, respectively (these parameters are the half widths of the corresponding transitions). The cross term \( W_{m\eta} \) is a rate of relaxation transition from state \( m \) to state \( \eta \). The parameter \( P \) is an incoherent pump rate of the \( \gamma \)-transition (we introduced it formally to have a population of the excited nuclear state; however, a real pump must start from other nuclear levels or even excite a different isomer). We define 

\[ C_{\eta_4} = S(\eta)(d_{\eta_4} \cdot E_\eta)/\hbar; \quad B_{\eta_3} = S(\eta)(d_{\eta_3} \cdot E_\eta)/\hbar, \]

where \( d_{\eta_3} = d_{\eta_4} = d_\eta \) is the matrix element (assumed to be real) of the dipole optical transition from the state \( \eta \) to the state \( 3 \) and \( d_{\eta_4} = d_{\eta_3} = d_\eta \) is the (real) matrix element for the \( \gamma \)-transition, \( E_\eta \) and \( E_\eta \) are the amplitudes of the laser-pump and \( \gamma \)-probe fields, respectively. The coefficients \( S(1) = -\sin \psi \) and \( S(2) = \cos \psi \) come from the wave function coefficients for the allowed \( \gamma \)-transitions because of level-mixing. The master equations contain the coherences 

\[ \sigma_{\eta_1} = \rho_{\eta_1} \exp[i(\Omega - \omega)t + i(K - K)z] \quad \text{and} \quad \sigma_{\eta_2} = \rho_{\eta_2} \exp[i(\Omega - \omega)t - i(K - K)z] \]

produced by the two-quantum transition involving the optical pump and the \( \gamma \)-probe. Decay of this coherence is caused by the coherence decay of the constituting transitions, i.e., \( \Gamma = \Gamma' + \Gamma_3 \).

As we consider the Mössbauer nuclei embedded in the crystal, there are certain constraints on the optical pump. First, at room temperature the line width of the optical transition (2\( \Gamma' \)) ranges from several cm\(^{-1}\) up to several hundreds cm\(^{-1}\) [35]. Because this homogeneous line broadening is caused by crystal phonons, a temperature decrease can reduce this broadening. However, there is a limit below which further cooling will reduce the number of particles participating in the resonant interaction. This limit can be estimated as 0.1 cm\(^{-1}\), i.e., several GHz, and set by the inhomogeneous broadening [36].

The excitation of only a small part of the inhomogeneous line will decrease the number of nuclei participating in the GWI. Because the nuclear cross section is already small, any additional reduction of the \( \gamma \)-interaction with nuclei is undesirable. The homogeneous width can be reduced only for weak optical transitions with small transition matrix elements. This is a second constraint in our model, limiting the value of the Rabi frequency which can be achieved by available laser sources. To simplify the notations we define the optical Rabi frequency \( R_\eta \) as related to the coefficient \( B_{\eta_3} \) in a following way 

\[ R_\eta = (d_\eta \cdot E_\eta)/\hbar. \]

Functions \( S(\eta) \) are considered as supplementary parameters dependent on tilting angle of the magnetic field.

Also for the \( \gamma \)-probe, the Rabi frequency, \( R_\gamma = (d_\gamma \cdot E_\gamma)/\hbar \), is smaller than the line width of the transition (\( R_\gamma < \Gamma' \)). However, while for the \( \gamma \)-transition the spontaneous decay rate of the excited state is comparable with the coherence decay, i.e., \( W_{\eta_4} = W_{\eta_3} = \Gamma_\eta \), the optical coherence decay is much faster than the spontaneous decay rate of the optically excited state because of phonon contribution [35], i.e., \( \Gamma \gg W_{\eta_3} \). Therefore, for the optical transition it is much simpler to reach the saturation, i.e., to satisfy the condition \( R_\eta > \sqrt{W_{\eta_3}/T} \), than to create Rabi splitting which takes place for \( R_\eta > \Gamma \). For this reason the Autler-Townes effect [37,38] will not play an important role in our scheme.

We study the stationary linear response to the \( \gamma \)-probe assuming that the sample is optically thin for \( \gamma \)-quanta. To calculate linear amplification coefficient within this approximation, one first derives the steady state solution \( \bar{p}_{\eta \eta}, \bar{\sigma}_{\eta \eta} \) of the master equations. Then evaluating the polarization corresponding to the steady state response to the \( \gamma \)-probe

\[
P(t) = \sum_{\eta=1,2} d_{\eta_4} \bar{\sigma}_{\eta_4} \exp(-i\Omega t + iKz) + \text{c.c.}
\]

one can use the following expression for the linear amplification coefficient for \( \gamma \)-quanta [39]

\[
\alpha_\gamma(\Omega) = -\frac{4\pi KN}{E_\gamma} \text{Im} \left( \sum_{\eta=1,2} d_{\eta_4} \bar{\sigma}_{\eta} \right),
\]

where \( N \) is the concentration of the Mössbauer nuclei in the sample, \( d_{\eta_4} \) and \( E_\gamma \) are the absolute
values of the corresponding vectors [the other symbols are defined after the master equations (6)-(13)]. To clarify the physical processes involved in the LM scheme, we express this coefficient as follows

\[
\alpha_g(\Omega) = \alpha_{14}(\Omega) + \alpha_{23}(\Omega) + \alpha_{34}(\Omega),
\]

(16)

where \( d_\gamma \) is a module of the vector \( d_\gamma \), \( \alpha_{\eta}(\Omega) \), with \( \eta = 1 \) or 2, is the contribution of the quantum transition \( \eta \rightarrow 4 \). It includes the branching via two paths, i.e., \( \eta \rightarrow 4 \) (the first path) and \( \eta \rightarrow \eta' \rightarrow 4 \) (the second path which comes from the coherence) with \( \eta' = 1 \) or 2. The latter path is represented by the term proportional to \( \tilde{c}_\eta \). The component \( \alpha_{14}(\Omega) \) represents the contribution of another branching involving the optical path \( \eta \rightarrow 3 \rightarrow 4 \) [see Eq. (10)].

Below we give a numerical example of GWI taking realistic values of the parameters. However, some of them may vary to a large extent. The dependence of the amplification coefficient \( \alpha_g(\Omega) \) on the frequency difference \( \Omega - \Omega_0 \) with \( \omega = \omega_0 \) is shown in Fig. 2. In this plot, the coefficient is normalized by the absolute value \( \alpha_0 \) of the absorption coefficient with zero laser and incoherent pumps. The order of magnitude of the parameters characterizing the nuclear transition, the optical transition and the relaxation of the spin sublevels is given below. We take the value of the ground state level splitting \( \omega_{21} = 1 \) kHz. The decay rate of the \( \gamma \)-coherence is \( \Gamma = 1 \) MHz. The half width at half maximum of the amplification line (see Fig. 2) coincides with this value. The relaxation rates of the nuclear transitions \( 4 \rightarrow 1 \) and \( 4 \rightarrow 2 \) are equal, \( W_{41} = W_{42} = 1 \) MHz. The decay rate of the optical coherence is taken as \( \Gamma = 3 \) GHz. This rate can be achieved at liquid nitrogen temperature [35, 36], if the phonon contribution is comparable with the inhomogeneous broadening caused by the crystal imperfections. The reduction of this rate is necessary as we have to satisfy the condition \( R_\gamma^2 \gg IT_{\delta M} \), which will

\[
\alpha_{14}(\Omega) = \operatorname{Re} \frac{4\pi KND_\gamma^2}{\hbar} \left( \tilde{c}_1 - \tilde{c}_3 \right) \sin^2\psi
\]

\[
+ \tilde{c}_1 \sin \psi \cos \psi,
\]

(17)

\[
\alpha_{23}(\Omega) = \operatorname{Re} \frac{4\pi KND_\gamma^2}{\hbar} \left( \tilde{c}_2 - \tilde{c}_3 \right) \cos^2\psi
\]

\[
+ \tilde{c}_2 \sin \psi \cos \psi,
\]

(18)

\[
\alpha_{34}(\Omega) = \operatorname{Re} \frac{4\pi KND_\gamma^2}{\hbar} \left( \frac{\sin^2\psi}{\Gamma_\gamma - i\Delta_1} \right)
\]

\[
+ \frac{\cos^2\psi}{\Gamma_\gamma - i\Delta_2} \left( \frac{R_\gamma}{\sigma_{34}} \right),
\]

(19)
be explained below. Also the condition $W_{3y} > \Gamma_M$ must be fulfilled. Therefore, we take $R_L = 10$ MHz, which is an order of magnitude higher than in the rotary echo experiment using cw excitation gated by an acousto-optical modulator [40]. For the spontaneous decay rate of the excited electron state we take $\Gamma_M = 150$ Hz. This is by no means an underestimated value, because hyperfine interaction creates a frozen core or diffusion barrier with suppressed spin flips of nearest neighbors [41,42]. For example, spin-echo decay of the frozen core nuclei, measured by the optical Raman heterodyne detection (Raman echo), has given $\sim 1$-msec dephasing time of the nuclear spin coherence [43]. It is worth to mention that in the experiment reported in [43], the first long laser pulse (10 msec) was applied before the rf pulses (forming an echo-inducing sequence) to create polarized nuclei in the frozen core. Without this polarization the echo signal was too weak to be detected. For the population difference of the ground state spin-sublevels we assume spin-lattice relaxation, which is slow, and we take $W_{12} = W_{21} = 40$ Hz [44]. The rate of incoherent nuclear pumping is

$$P = 100 \text{ kHz.}$$

This rate is enough to transfer 5% of the total population to the nuclear excited state [4]. Laser pumping satisfying the conditions $\omega = \omega_0$ and $R_L^2 \gg I \Gamma_M$ does not excite appreciably the state $|3\rangle$. For example, with the parameters listed above, the population of the optically excited state $|3\rangle$ is only 2.6% of the total. This small excitation, even for large laser powers, is caused by the population trapping in a dark state, which is similar to the nonabsorption resonances observed in sodium vapor [45–48].

In our case the optical pump changes the density matrix of the impurity as follows

$$2 \bar{\rho}_{11} = 2 \bar{\rho}_{22} = 0.928,$$

$$\bar{\rho}_{12} + \bar{\rho}_{21} = 0.871,$$

$$\bar{\rho}_{31} = -i(\bar{\rho}_{12} - \bar{\rho}_{21}) = 0.026,$$

$$\bar{\rho}_{44} = 0.046,$$

where the condition $\sum_{m=1}^4 \bar{\rho}_{mm} = 1$ is imposed. The population of the ground states is reduced from 95% down to 92.8% due to the laser pump, transferring 2.6% of the population to the state $|3\rangle$. Because of the ground states population reduction, the nuclear excited state population is also reduced since $\bar{\rho}_{44} = 0.05(\bar{\rho}_{11} + \bar{\rho}_{22})$. This expression comes from the incoherent pumping starting from the ground states

$$\bar{\rho}_{44} = \frac{P}{W_{41} + W_{42}} (\bar{\rho}_{11} + \bar{\rho}_{22}).$$

Taking into account that $\bar{\rho}_{11} = \bar{\rho}_{22}$, one can rewrite Eq. (16) as follows

$$\alpha_y(\Omega_0) = \alpha_0 \left[ 2 \bar{\rho}_{44} - (\bar{\rho}_{11} + \bar{\rho}_{22}) + (\bar{\rho}_{12} + \bar{\rho}_{21}) \right. + i(\bar{\rho}_{12} - \bar{\rho}_{21}) \frac{\omega_{21}}{2 \Gamma_y} \right],$$

where the term $\alpha_y(\Omega_0)$ is omitted, as it is small. One can also disregard the last term in the square brackets of Eq. (25) since $\omega_{21}/2 \Gamma_y = 0.5 \times 10^{-3}$ is very small. Then the substitution of the density matrix elements from Eqs. (20)–(23) into Eq. (25) gives $\alpha_y(\Omega_0) = 0.036 \alpha_0$. This means that, due to the interference of the quantum paths, almost 40% of the excited state population contributes to the stimulated $\gamma$-emission and the rest is quenched by the
from the reduced master equations state matrix elements in Eq. 25 can be calculated. The low-frequency splitting, $G_{WI}$, we consider first the simplified case of zero resonance absorption because of the imperfect match of these paths. This mismatch comes from the decay of the low-frequency coherence $\bar{n}_{12}$ with the rate $\Gamma_{34}$. Also the shift of the laser pump frequency $\omega$ from the value $\omega_0$ produces an unbalance of quantum paths. The dependence of the density matrix elements, which are involved in the coefficient $\alpha_s(\Omega_0)$, on $\omega - \omega_0$ is shown in Fig. 3. The coherence, represented by $\bar{n}_{12} + \bar{n}_{21}$, is decreased by 2 for $\omega - \omega_0 = 5\Gamma$.

4. Analysis of the analytical solution

To explain the physical processes involved in GWI, we consider first the simplified case of zero low-frequency splitting, $\omega_{21} = 0$. Then the steady state matrix elements in Eq. (25) can be calculated from the reduced master equations

$$\dot{n}_{12} = -2W_r \rho_{44} + Pn_{12} ,\quad (26)$$

$$\dot{n}_{33} = G(n_{12} - u_{12}) - 2(W_L + G) \rho_{33} ,\quad (27)$$

$$\dot{u}_{12} = -\Gamma_{34} u_{12} + G(n_{12} - u_{12}) - 2G\rho_{33} ,\quad (28)$$

$$\dot{u}_{12} = -\Gamma_{34} u_{12} + G(n_{12} - u_{12}) - 2G\rho_{33} ,\quad (29)$$

where $n_{12} = \rho_{11} + \rho_{22}$, $u_{12} = \rho_{12} + \rho_{34}$, $W_r = W_{41} = W_{42}$, $W_L = W_{31} = W_{32}$ and $G = \frac{R_L^2}{\Gamma}$. These master equations are derived from Eqs. (6)–(13) under the following conditions. First, we set $C_{n4} = 0$, the condition which corresponds to zero-order approximation for the probe field. Second, the contribution of the coherence $\sigma_{34}$ is neglected. Third, level mixing satisfies the condition $\psi = \pi/4$ and fourth, $\Gamma \gg R_L$, which allows us to use the approximation of wide optical line where it is possible to neglect the derivative in Eq. (11) [49]. The latter procedure corresponds to the adiabatic elimination of the optical coherence.

In terms of the parameters

$$\hat{\Gamma}_M = \frac{\Gamma_M}{1 + \frac{\Gamma_M}{G}} ,\quad (30)$$

$$\Phi = \frac{\hat{\Gamma}_M / \Gamma_M}{P} ,\quad (31)$$

$$p = \frac{P}{2W_r} ,\quad (32)$$

the stationary solution of Eqs. (26)–(29), satisfying the condition $n_{12} + \rho_{33} + \rho_{44} = 1$, is

$$\bar{n}_{12} = \frac{\hat{\Gamma}_M}{(3 + 2p) \hat{\Gamma}_M + (2 + 2p)W_L} ,\quad (34)$$

$$\bar{n}_{33} = \frac{2(W_L + \hat{\Gamma}_M)}{(3 + 2p) \hat{\Gamma}_M + (2 + 2p)W_L} ,\quad (35)$$

$$\bar{u}_{12} = \Phi \cdot \frac{2W_L}{(3 + 2p) \hat{\Gamma}_M + (2 + 2p)W_L} .\quad (36)$$

We see that trapping in the dark state ($\bar{n}_{12} \sim 1$, $\bar{n}_{33} \ll 1$) takes place if $W_L \gg \hat{\Gamma}_M$. Because $\hat{\Gamma}_M$ depends on $\Gamma_M$ and $G$, one can conclude that the particle is captured in the dark state by spontaneous emission from state $|3\rangle$, if the spontaneous decay rate, $W_L$, of this state is larger than the low-frequency coherence decay $\Gamma_M$. This happens at the excitation level satisfying the condition $G \gg \Gamma_M$. The former condition ($W_L \gg \hat{\Gamma}_M$) relates to the sample and cannot be adjusted by external means, whereas the latter ($G \gg \Gamma_M$) specifies only the intensity of the laser pump necessary to achieve this trapping. If the first
condition is not fulfilled, then no pump will lead to trapping.

The same conclusion can be drawn from the analysis of the GWI condition. Amplification takes place if \( \alpha_\gamma(\Omega_0) \) is positive [see Eq. (25)], i.e., if

\[ 2\pi_{14} - \pi_{12} + \pi_{12} > 0. \]  \hspace{1cm} (37)

The latter inequality is reduced to

\[ \frac{P}{W_{y} - P} > \frac{\Gamma M + \Gamma T M}{W_L + R_L^2}, \]  \hspace{1cm} (38)

where one can recognize both conditions discussed above. The left hand side of the inequality is supposed to be smaller than one, because we consider amplification without inversion \( (P \ll W_L) \). Then, to satisfy the necessary condition (38), two inequalities, i.e. \( W_L > \Gamma M \) and \( R_L^2 > \Gamma T M \), must be fulfilled simultaneously.

If \( \omega_{21} \neq 0 \) and \( \omega = \omega_0 \), the stationary solution of the master equation keeps the same form (33)–(36), except for the values of \( \Gamma M \) and \( \Phi \), which are modified as follows

\[ \Gamma_M = \Gamma_M + \frac{a}{1 + 2b + c}, \]  \hspace{1cm} (39)

\[ \Phi = \frac{1 + b}{1 + 2b + c}. \]  \hspace{1cm} (40)

where

\[ a = \frac{\Gamma M^2 + \omega_{21}^2 \omega_{21}}{R_L^2}, \]  \hspace{1cm} (41)

\[ b = \frac{2\Gamma T M - \omega_{21}^2}{2R_L^2}, \]  \hspace{1cm} (42)

\[ c = \frac{4\Gamma^2 + \omega_{21}^2(\Gamma_M^2 + \omega_{21}^2)}{4R_L^2}. \]  \hspace{1cm} (43)

We consider the case where \( \omega_{21} \) is always much smaller than \( R_L \). Therefore, in Eq. (39) the contribution of \( a \) is dominant compared to \( b \) and \( c \). In this domain the effective decay rate \( \Gamma_M \) increases with the increase of the low-frequency splitting \( \omega_{21} \) (see Fig. 4). The change is small if \( \omega_{21} < R_L^2 \Gamma_M / T \). We emphasize that the limiting value \( R_L^2 \Gamma_M / T \) is much larger than \( \Gamma_M \). Therefore, the nuclear spin states 1 and 2 are not lumped by the dephasing \( \Gamma_M \) and \( \omega_{21} \), can even be measured by NMR techniques. With the increase of the parameter \( \Gamma_M \), the population of the dark state or trapping decreases and the amplification condition becomes hard to fulfill. The latter is modified as follows

\[ \frac{P}{W_y - P} > \frac{\Gamma M}{W_L \Phi} + \frac{1 - \Phi}{\Phi}. \]  \hspace{1cm} (44)

The dependence of the amplification coefficient on \( \omega_{21} \), with \( \omega = \omega_0 \) and \( \Omega = \Omega_0 \), is shown in Fig. 5. The parameters of the transition and the excitation are the same as before. The plot shows that, if the

![Fig. 4. Dependence of the modified decay rate \( \Gamma_M \) (solid line, see Eq. (39)) and the function \( \Phi \) (dashed-dotted line, see Eq. (40)), on the frequency difference of the spin levels \( \omega_{21} \). The function \( \Phi \) is shown for comparison. It decreases by about 8% in a frequency range of 10 kHz. We take the following parameters of the system \( \omega = \omega_0 \), \( \Gamma = 3 \) GHz, \( R_L = 10 \) MHz, \( \Gamma_M = 150 \) Hz.](image)

![Fig. 5. Dependence of the amplification coefficient \( \alpha_\gamma / \alpha_0 \) on the spin-splitting frequency \( \omega_{21} \) for \( \omega = \omega_0 \) and \( \Omega = \Omega_0 \). The parameters of the transition \( (\Gamma, \Gamma_M) \) and the laser excitation \( (R_L) \) are the same as in Fig. 4. and \( W_L = 3 \) kHz, \( P = 100 \) kHz. The parameter \( \alpha_0 \) is defined at the value \( \omega_{21} = 0 \).](image)
low-frequency $\omega_{21}$ exceeds 2.3 kHz, the coefficient $\alpha_z(Q_0)$ changes sign as the gain condition (44) is violated. The physical reason of this violation can be interpreted within the spin-vector model. The optical pump creates the coherence between states $|1\rangle$ and $|2\rangle$, which corresponds to the build up of the magnetization $M$ along the laser beam ($z$-axis). This magnetization is transverse to the magnetic field $H_z$. Therefore, the latter creates a torque $C = M \times H_z$ resulting in a decrease of the magnetization $M$. The competition of the pump and the force induced by the field $H_z$ may result in a decrease of the magnetization $M$. As the spin leaves the dark state with the rate $\omega_{21}$, the increase of the low frequency splitting will decrease the population of the dark state.

5. Conclusion

We considered the case when $\gamma$-absorption for a particular transition can be essentially reduced. This becomes possible if we make two ground state spin levels close in energy space. The crossing and mixing of them allows splitting and interference of the quantum paths within one $\gamma$-emission (absorption) line. Indirect optical pumping of the nuclear spin states via electro-nuclear interaction is capable to create the coherence of the mixed states. This coherence results in the suppression of the $\gamma$-absorption because of the destructive interference of the quantum paths. The emission for the same $\gamma$-transition is not quenched as it terminates in another superposition of the mixed states and the interference of the quantum paths is constructive. So, just by the state mixing we can avoid bichromatic excitation and emission in order to obtain GWI. This is important as simultaneous excitation of several transitions is hard to expect for the $\gamma$-domain.

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