

Steady-state solutions of the density matrix equations for multiple resonance experiments

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Analytical steady-state solutions of the density matrix equation for a spin or an atomic multiplet subjected to two or three electromagnetic fields near resonance are obtained within the semiclassical approach, in the nondegenerate case. In comparison with other treatments, these solutions do not assume simplified approximations concerning the number of levels, or the relative magnitudes of the Rabi frequencies and relaxation rates. Consequently, they contain all possible interference effects among single and multiple quantum absorption and emission processes, and parametric processes. The solutions are useful not only for double or triple resonance experiments, they can be used to find the conditions that are necessary to attain optimum quantum amplification and detection, or maximum frequency conversion efficiencies and resonant enhancements corresponding to three or four wave mixing processes.

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1. Introduction

Conventional magnetic resonance spectroscopy, or laser spectroscopy follow the simplest scheme of the experiment, where a coherent electromagnetic field near resonance acts upon the sample. Developed techniques use a multiple frequency irradiation. Experiments with multiple irradiation of the sample are carried out in nuclear and electron magnetic resonance, laser spectroscopy, quantum amplification and detection, frequency conversion, etc. There are two kinds of such experiments. a) A multilevel system is subjected to two or three electromagnetic fields near resonance so that every transition involves a different pair of levels: electron-nuclear double resonance (ENDOR), electron-electron double resonance (ELDOR), optical detected magnetic resonance (ODMR), triple resonance [1], lasing [2,3] or masing [4,5] with or without inversion, nonlinear optical processes using electromagnetically induced transparency [6], resonant enhancement of the parametric frequency processes [7], etc. b) A two-level system is subjected to many electromagnetic fields so that the incident frequencies are within the resonance linewidth. Thus longitudinally detected electron spin resonance (LODESR) [8], or continuous wave multiwave EPR (CWMQEP) [9] uses bichromatic microwave resonant irradiation, while others use polychromatic resonant irradiation [10].

The purpose of this work is to obtain analytical steady-state solutions of the density matrix equation for a spin, or an atomic multiplet subjected to two or three electromagnetic fields near resonance. These solutions are obtained within the semiclassical approach and are valid for experiments of type a, in the nondegenerate case. In comparison with other treatments [1, 3, 6, 7, 11-13], these solutions do not assume simplified approximations concerning the number of levels, or the relative magnitudes of the Rabi frequencies and relaxation rates.

Consequently, they contain all possible interference effects among single and multiple quantum absorption and emission processes, and parametric processes.

2. The electromagnetic powers absorbed, emitted or dispersed by the sample

The treatment is similar to that used in other papers [5,14-18], where the fields are exactly at resonance. Considering the rotating wave approximation, the powers

$P_{kj}^* < 0$ absorbed, $P_{kj}^* > 0$ emitted, and P_{kj}^d dispersed by a multilevel spin (or atomic) system subjected to many quasi-monochromatic fields close to resonance are:

$$P_{kj}^{*,d} = 2(E_k - E_j)N\Omega_{kj}^{*,d} \quad (1)$$

where N is the total number of spins (atoms), and E_i the

energies of the involved levels. $2\Omega_{kj}^{*,d}$ may be interpreted as the number of photons of angular frequency

$\omega_{kj} \equiv \frac{(E_k - E_j)}{\hbar}$ absorbed, emitted or dispersed by an

atom in one second. ω_{kj} are the frequencies of the involved fields considered near resonance with respect to

the pair of levels k, j . Any ratio of these frequencies is supposed to be not an integer number (the nondegenerate case). The populations N_i of the levels are given by the

solutions for the diagonal elements $\rho_{ii} = \frac{N_i}{N}$ of the density matrix [5,14]:

$$\rho_{ii} = \rho_{ii}^0 + 2 \sum_{k,j} K_{kj}^i \Omega_{kj}^*; k < j \quad (2)$$

where ρ_{ii}^0 are the thermal equilibrium values of ρ_{ii} . The spin-lattice relaxation times (or the population decay times) K_{kj}^i are given by

$$\sum_{i \neq m} (-K_{mn}^m w_{mi} + K_{mn}^i w_{im}) = \sum_{i \neq n} (K_{mn}^n w_{ni} - K_{mn}^i w_{in}) = 1 \quad (3a)$$

$$\sum_{i \neq j} (K_{mn}^j w_{ji} - K_{mn}^i w_{ij}) = 0 \quad (j \neq m, n) \quad (3b)$$

where $w_{jn}/w_{nj} = \rho_{nn}^0/\rho_{jj}^0$. w_{ij} are the spin-lattice relaxation rates (or the population decay rates) characterising the pair of levels i, j . These spin-lattice relaxation times K_{kj}^i exhibit the following properties

$$\sum_i K_{mn}^i = 0; \quad K_{mn}^i = -K_{nm}^i; \quad K_{nm}^i = K_{n\sigma}^i + K_{\sigma m}^i \quad (4)$$

Note that the solution (2) is valid for any level of the multiplet, involved or not in the transitions.

Ω_{kj}^* and Ω_{kj}^d are the imaginary and real parts of

$$\Omega_{kj} = \int q_{kj}(\omega_{kj}) \rho_{kj}(\omega_{kj}) D_{kj}(\omega) d\omega, \quad \text{where}$$

$$\begin{aligned} & \left[(T_2^{mn})^{-1} (1 + \Delta_{mn}^2) + \left(\frac{1 + \Delta_{mn} \Delta_{\sigma m}^*}{1 + \Delta_{\sigma m}^{*2}} \right) \langle p_{n\sigma}^2 \rangle T_2^{\sigma m} + \left(\frac{1 + \Delta_{mn} \Delta_{n\sigma}^*}{1 + \Delta_{n\sigma}^{*2}} \right) \langle p_{\sigma m}^2 \rangle T_2^{n\sigma} \right] \Omega_{mn}^* - \\ & - \left[\left(\frac{\Delta_{mn} - \Delta_{\sigma m}^*}{1 + \Delta_{\sigma m}^{*2}} \right) \langle p_{n\sigma}^2 \rangle T_2^{\sigma m} + \left(\frac{\Delta_{mn} - \Delta_{n\sigma}^*}{1 + \Delta_{n\sigma}^{*2}} \right) \langle p_{\sigma m}^2 \rangle T_2^{n\sigma} \right] \Omega_{mn}^d - \left(\frac{1 + \Delta_{mn} \Delta_{\sigma m}^*}{1 + \Delta_{\sigma m}^{*2}} \right) \langle p_{mn}^2 \rangle T_2^{\sigma m} \Omega_{n\sigma}^* + \\ & + \left(\frac{\Delta_{mn} - \Delta_{\sigma m}^*}{1 + \Delta_{\sigma m}^{*2}} \right) \langle p_{mn}^2 \rangle T_2^{\sigma m} \Omega_{n\sigma}^d - \left(\frac{1 + \Delta_{mn} \Delta_{n\sigma}^*}{1 + \Delta_{n\sigma}^{*2}} \right) \langle p_{mn}^2 \rangle T_2^{n\sigma} \Omega_{\sigma m}^* + \left(\frac{\Delta_{mn} - \Delta_{n\sigma}^*}{1 + \Delta_{n\sigma}^{*2}} \right) \langle p_{mn}^2 \rangle T_2^{n\sigma} \Omega_{\sigma m}^d = \\ & = (\rho_{mm} - \rho_{nn}) \langle p_{mn}^2 \rangle + (\rho_{mm} - \rho_{\sigma\sigma}) T_2^{\sigma m} \left[\left(\frac{1 + \Delta_{mn} \Delta_{\sigma m}^*}{1 + \Delta_{\sigma m}^{*2}} \right) \alpha_{n\sigma m}^{\sin} + \left(\frac{\Delta_{mn} - \Delta_{\sigma m}^*}{1 + \Delta_{\sigma m}^{*2}} \right) \alpha_{n\sigma m}^{\cos} \right] + \\ & + (\rho_{nn} - \rho_{\sigma\sigma}) T_2^{n\sigma} \left[\left(\frac{1 + \Delta_{mn} \Delta_{n\sigma}^*}{1 + \Delta_{n\sigma}^{*2}} \right) \alpha_{n\sigma m}^{\sin} + \left(\frac{\Delta_{mn} - \Delta_{n\sigma}^*}{1 + \Delta_{n\sigma}^{*2}} \right) \alpha_{n\sigma m}^{\cos} \right] \end{aligned} \quad (5)$$

and other two equations obtained by circular permutations of mn , $n\sigma$, and σm . There are also other three equations by replacing in (5) Ω_{kj}^* by Ω_{kj}^d , Ω_{kj}^d by $-\Omega_{kj}^*$,

$\alpha_{n\sigma m}^{\sin}$ by $-\alpha_{n\sigma m}^{\cos}$, $\alpha_{n\sigma m}^{\cos}$ by $\alpha_{n\sigma m}^{\sin}$, and $(\rho_{mm} - \rho_{nn}) \langle p_{mn}^2 \rangle$ by $\Delta_{mn}(\rho_{mm} - \rho_{nn}) \langle p_{mn}^2 \rangle$.

$\langle p_{kj}^2 \rangle = \bar{p}_{kj}^2 = \int p_{kj}^2(\omega_{kj}) D_{kj}(\omega) d\omega$ are the mean square values of the Rabi frequencies. $\Delta_{kj} = \frac{\Delta\omega_{kj}}{R_{kj}}$, where

$\Delta\omega_{kj} = -\Delta\omega_{jk}$ are the frequency detunings of the three

$\rho_{kj}(\omega_{kj})$ are the off-diagonal elements of the density matrix, and $q_{kj}(\omega_{kj}) = p_{kj}(\omega_{kj}) e^{-i[\omega_{kj}t + \varphi_{kj}(\omega_{kj})]}$. If the Rabi frequencies p_{kj} that represent the interactions of the system with the electromagnetic fields are real, $\varphi_{kj}(\omega_{kj})$ represents the phase of one wave of frequency $\omega_{kj} = \omega_{kj}^0 + \delta\omega_{kj}$, ω_{kj}^0 being the frequency of a quasi-monochromatic field. In other words, we consider that the spectral distribution $D_{kj}(\omega)$ is non-vanishing for $|\delta\omega_{kj}| \ll (T_2^{kj})^{-1}$, where $(T_2^{kj})^{-1} = -R_{kj}$ is the linewidth of a homogeneously broadening simple transition between the levels k and j .

3a. Double or triple resonance

Within the treatment mentioned above, in the case of triple resonances, the solutions for the power parameters

Ω_{kj}^{*d} are obtained by solving a system of six coupled equations:

quasi-monochromatic fields from the resonance:

$$\Delta\omega_{kj} = \frac{E_k - E_j}{\hbar} - \omega_{kj}^0.$$

$$\Delta_{\sigma m}^* = \frac{\Delta\omega_{\sigma m}^*}{R_{\sigma m}} = \frac{\Delta\omega_{\sigma m} - \Delta\omega_{mn}}{R_{\sigma m}}; \quad (6)$$

and other two similar equations for $\Delta_{n\sigma}^*$ and Δ_{mn}^* . Note that the frequencies of the three involved fields are not necessary correlated so that

$$\omega_{nm}^0 = \omega_{n\sigma}^0 + \omega_{\sigma m}^0 \quad (7).$$

Thus, if the condition (7) is not satisfied, in Eq.(6)

$$\Delta\omega_{kj}^* \neq \Delta\omega_{kj}.$$

$$\alpha_{nom}^{sin} = \sum_{\omega_{nm}} p_{mn}(\omega_{nm}) \{ \sum_{\sigma} p_{n\sigma}(\omega_{n\sigma}) p_{om}(\omega_{om}) \sin[\varphi_{nm}(\omega_{nm}) - \varphi_{n\sigma}(\omega_{n\sigma}) - \varphi_{om}(\omega_{om})] \} \quad (8)$$

where the subscript " " means that the summation is made all over the frequencies so $\omega_{n\sigma} + \omega_{om} = \omega_{nm}$, and where p_{kj} are real. α_{nom}^{cos} is defined similarly by replacing the function $\sin$ with $\cos$. Formally, we can write $\alpha_{nom}^{sin} = \langle p_{nom}^3 \rangle \langle \sin \Delta\varphi^* \rangle$ and

$$\alpha_{nom}^{cos} = \langle p_{nom}^3 \rangle \langle \cos \Delta\varphi^d \rangle$$

where

$$\langle p_{nom}^3 \rangle = \sum_{\omega_{nm}} p_{nm}(\omega_{nm}) [\sum_{\sigma} p_{n\sigma}(\omega_{n\sigma}) p_{om}(\omega_{om})]$$

represents the frequency overlapping of the three fields, while $\Delta\varphi^{*,d}$ defines the phase mismatch for absorption and dispersion, respectively. $\Delta\varphi^* = \Delta\varphi^d$ and $\langle p_{nom}^3 \rangle$ reaches its maximum value in the ideal case of three strictly monochromatic fields, whose frequencies are perfectly correlated. Therefore, the last terms in Eqs. (5) makes a clear distinction between the ideal monochromatic case, and the quasi-monochromatic case.

Note that the terms proportional to α_{nom}^{sin} , and α_{nom}^{cos} which correspond to the coherent mixing of the fields are nonvanishing as long as their frequencies are correlated, or

the condition (7) is satisfied. In this case $\Delta_{kj}^* = \Delta_{kj}$.

The system of six coupled Eqs. (5) corresponds to a triple resonance if the three Rabi frequencies p_{mn} , p_{om} and $p_{n\sigma}$ are nonvanishing. The simple or double resonance case is obtained if two or one among the above Rabi frequencies are vanishing, respectively.

3b. Triple resonance with correlated frequencies

We must emphasize that concerning the triple resonance with correlated frequencies (see Eq.(7)), there is an important difference between a spin multiplet and an atomic multiplet. In the case of spin multiplets, all the three Rabi frequencies corresponding to magnetic dipole transitions could be simultaneously nonvanishing [5, 14]. In the case of atomic multiplets, for systems without inversion centre, the three Rabi frequencies, corresponding to electric-dipole transitions, may not be simultaneously nonvanishing and a three wave mixing process is not possible. In this case, one among the three Rabi frequencies in Eqs.(5) corresponds to a two phonon transition and we deal with a four-wave mixing process [3,11-13].

If the frequencies of the involved fields are correlated, or the condition (7) is fulfilled, the system (5) of six

coupled equations are easily separated in two sets of three

coupled equations for Ω_{kj}^* and Ω_{kj}^d respectively.

For example the solutions for the power parameters corresponding to the absorbed or emitted fields become:

$$\begin{aligned} \Omega_{mn}^* S_{mn} \Delta + \frac{\Delta_{mn,n\sigma}}{S_{n\sigma}} [\langle p_{om}^2 \rangle \Omega_{mn}^* - \langle p_{mn}^2 \rangle \Omega_{om}^*] + \frac{\Delta_{mn,om}}{S_{om}} [\langle p_{n\sigma}^2 \rangle \Omega_{mn}^* - \langle p_{mn}^2 \rangle \Omega_{n\sigma}^*] = \\ = (\rho_{mn} - \rho_{mn}) \langle p_{mn}^2 \rangle \Delta + \frac{\langle p_{mn}^2 \rangle \langle p_{n\sigma}^2 \rangle}{S_{om}} (\Delta_{mn} - \Delta_{om}) \left(\frac{\rho_{mn} - \rho_{mn}}{S_{mn}} \Delta_{mn} - \frac{\rho_{n\sigma} - \rho_{mn}}{S_{n\sigma}} \Delta_{n\sigma} \right) + \\ + \frac{\langle p_{mn}^2 \rangle \langle p_{om}^2 \rangle}{S_{n\sigma}} (\Delta_{mn} - \Delta_{n\sigma}) \left(\frac{\rho_{mn} - \rho_{mn}}{S_{mn}} \Delta_{mn} - \frac{\rho_{n\sigma} - \rho_{n\sigma}}{S_{n\sigma}} \Delta_{n\sigma} \right) + \\ + \left[\frac{\Delta_{mn,om}}{S_{om}} (\rho_{mn} - \rho_{n\sigma}) + \frac{\Delta_{mn,n\sigma}}{S_{n\sigma}} (\rho_{mn} - \rho_{n\sigma}) \right] \alpha_{nom}^{sin} + \\ + \left[\frac{\Delta_{mn} - \Delta_{om}}{S_{om}} (\rho_{mn} - \rho_{n\sigma}) + \frac{\Delta_{mn} - \Delta_{n\sigma}}{S_{n\sigma}} (\rho_{mn} - \rho_{n\sigma}) \right] \alpha_{nom}^{cos} + \\ + \frac{\Delta_{om} - \Delta_{n\sigma}}{R_{mn} S_{n\sigma} S_{om}} [\langle p_{mn}^2 \rangle (\rho_{mn} - \rho_{mn}) \langle p_{mn}^2 \rangle + (\rho_{mn} - \rho_{n\sigma}) \langle p_{n\sigma}^2 \rangle + (\rho_{n\sigma} - \rho_{mn}) \langle p_{om}^2 \rangle] \alpha_{nom}^{cos} \end{aligned} \quad (9)$$

and other two similar equations obtained by circular permutations, where

$$\Delta = 1 + \frac{\langle p_{mn}^2 \rangle (1 + \Delta_{n\sigma} \Delta_{om})}{S_{n\sigma} S_{om}} + \frac{\langle p_{n\sigma}^2 \rangle (1 + \Delta_{mn} \Delta_{om})}{S_{mn} S_{om}} + \frac{\langle p_{om}^2 \rangle (1 + \Delta_{n\sigma} \Delta_{mn})}{S_{n\sigma} S_{mn}} \quad (10)$$

$$\begin{aligned} \Delta_{mn,n\sigma} = \Delta_{n\sigma,mn} = 1 + \Delta_{mn} \Delta_{n\sigma} + \frac{\langle p_{mn}^2 \rangle}{R_{n\sigma} R_{om}} + \frac{\langle p_{n\sigma}^2 \rangle}{R_{mn} R_{om}} + \frac{\langle p_{om}^2 \rangle}{R_{n\sigma} R_{mn}} + \\ + \frac{\Delta_{om}}{S_{om}} \left[\frac{\langle p_{mn}^2 \rangle}{R_{n\sigma}} (\Delta_{mn} - \Delta_{om}) + \frac{\langle p_{n\sigma}^2 \rangle}{R_{mn}} (\Delta_{n\sigma} - \Delta_{om}) \right] \end{aligned} \quad (11)$$

$$\text{and } S_{kj} = R_{kj} (1 + \Delta_{kj}^2).$$

The solutions (2), (5), and (9) are valid within the rotating wave approximation, as long as the

inhomogeneous broadening is negligible, $\bar{P}_{ij} \ll \omega_{ij}^o$, and for a spin multiplet [16]

$$\langle p_{kj}^2(\omega_{mn}) \rangle / (R'_{rs} R_{kj}) \ll 1 \quad (12)$$

$-R'_{rs}$ are relaxation rates corresponding to a pair of different vibrational states and represent the linewidth of the infrared spectra. It is very large (of tens cm^{-1} or about 10^4 - 10^5 mT) in comparison with the Lorentzian EPR linewidth $-R_{kj}$. Consequently, these solutions are valid not only for weak fields, but also for very strong electromagnetic fields whose magnetic amplitudes of the cavity modes are of a few or tens of mT. In this case the Rabi frequency p_{kj} could be comparable or even higher than $-R_{kj}$. Thus unlike other treatments [1], the Eqs.(5) do not assume any other simplified approximation.

Within the rotating wave approximation, only one between the two counter-rotating circular polarized fields of the same frequency has an important contribution to the solutions (5), and (9). However if the three involved fields

have their frequencies correlated, the solutions (9) correspond to an important simplification because Δ in Eqs. (9) is simplified. So it is possible that the rotating wave approximation, and consequently the solutions (9) to be not valid for $\Delta=0$. For example, let us consider the simplest case of a single field of frequency ω_{nm}^0 applied near resonance. In this case $\langle p_{n\sigma}^2 \rangle$, $\langle p_{\sigma m}^2 \rangle$, $\Omega_{n\sigma}^*$, and $\Omega_{\sigma m}^*$ in Eq.(9) are vanishing, and this equation becomes:

$$\Omega_{mn}^* R_{mn} \Delta = (\rho_{nn} - \rho_{mm}) \langle p_{mn}^2 \rangle \Delta \quad (13)$$

and the solution for Ω_{mn}^* corresponds to a second simplification by Δ . Considering Eqs. (1), (2), (3), and (13), one obtains for the electromagnetic power absorbed by the sample the usual result

$$P_{mn}^* = \frac{2(E_m - E_n)N(\rho_{mm}^0 - \rho_{nn}^0) \langle p_{mn}^2 \rangle (T_2^{mn})^{-1}}{\Delta \omega_{mn}^2 + (T_2^{mn})^{-2} + 2 \langle p_{mn}^2 \rangle (T_2^{mn})^{-1} T_1^{mn}} \quad (14)$$

where $-R_{mn} = (T_2^{mn})^{-1}$ is the spin-spin relaxation rate (or the dephasing rate), and $T_1^{mn} = K_{mn}^n - K_{mn}^m$ is the spin-lattice relaxation time (or the population decay time) corresponding to the pair of levels m, n . Let us take for simplicity the applied field exactly at resonance $\Delta \omega_{nm} = \Delta \omega_{n\sigma} + \Delta \omega_{\sigma m} = 0$. Consequently $\Delta \omega_{n\sigma} = -\Delta \omega_{\sigma m} = \Delta \omega$, and considering in Eq.(10) $\Delta=0$ one obtains:

$$1 + \frac{\langle p_{mn}^2 \rangle \left(1 - \frac{\Delta \omega^2}{R_{n\sigma} R_{\sigma m}} \right)}{R_{n\sigma} R_{\sigma m} \left(1 + \frac{\Delta \omega^2}{R_{n\sigma}^2} \right) \left(1 + \frac{\Delta \omega^2}{R_{\sigma m}^2} \right)} = 0 \quad (15)$$

The solutions of the Eq.(15) are

$$(\Delta \omega^2)_{1,2} = \frac{1}{2} \left[\langle p_{mn}^2 \rangle (-R_{\sigma m}^2 - R_{n\sigma}^2) \pm \left(\langle p_{mn}^2 \rangle^2 - 2(R_{\sigma m} + R_{n\sigma})^2 \langle p_{mn}^2 \rangle + (R_{\sigma m}^2 - R_{n\sigma}^2)^2 \right)^{\frac{1}{2}} \right] \quad (16)$$

Consider for simplicity the usual case:

$$-R_{\sigma m} = -R_{n\sigma} = (T_2)^{-1}. \text{ Consequently if}$$

$$\langle p_{mn}^2 \rangle \geq 8(T_2)^{-2} \quad (17)$$

there are 2, or 4 solutions for the frequency detuning $\Delta \omega$ from the resonance of the quasi-monochromatic so that $\Delta=0$:

$$(\Delta \omega^2)_{1,2} = \frac{1}{2} \left[\langle p_{mn}^2 \rangle - 2(T_2)^{-2} \pm \left(\langle p_{mn}^2 \rangle^2 - 8(T_2)^{-2} \langle p_{mn}^2 \rangle \right)^{\frac{1}{2}} \right] \quad (18)$$

We have to emphasize that if the applied microwave field is strong enough and the condition (17) is fulfilled and there is a noise signal of frequency $\omega_{n\sigma}$, or $\omega_{\sigma m}$, the solutions of the Eqs.(9) could ceased to be valid for frequency detunings given by Eqs.(16), or (18).

4. Conclusions

The solutions of the density matrix equation given in this work are useful not only for double or triple resonance experiments, since they can be used to find the conditions that are necessary to attain maximum frequency conversion efficiencies and resonant enhancements of the parametric processes corresponding to three (of four) field mixing processes [5, 17,18].

It should be noted that, as long as the photo-ionization is negligible, these solutions are valid for any atomic multiplet. So it is possible to perform a comparison between the microwave case and the optical case.

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