

Four-photon parametric interaction under the condition of magnetic sublevel coherence

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The influence of the atom magnetic sublevel coherence on the nondegenerate four-wave parametric light scattering has been theoretically investigated taking into account the multiplet structure of the excited state. The theory is based on the irreducible tensorial formalism that allows us to take into consideration the relaxation related to both the nonuniform population and Zeeman coherence between the magnetic sublevels. The coefficient of the wave parametric coupling has been studied and three physical mechanisms (normal population, cross population and Zeeman coherence) are discussed. The results can be used, in particular, in high-resolution spectroscopy of the dipole-forbidden transitions.

In recent years, the influence of magnetic sublevel coherence on nonlinear phenomena in complex quantum systems has been investigated. Among these phenomena, the ones connected with nonlinear population and Zeeman coherence play special role, having wide applications in high-resolution spectroscopy [1] – [3]. Such interference effects as Hanle effect, level-crossing, quantum beating, population trapping have been intensively studied [4].

In this paper, we have developed a polarization theory of four-wave mixing in multilevel resonant system taking into account the magnetic sublevel coherence. Since the excited state of alkali metal atoms has a doublet structure, the theoretical investigation of the light resonance interaction has to consider both the third level [5] and the degeneracy of the levels. The theory is based on the irreducible tensorial formalism that allows us to take into account the relaxation related to the nonuniform population and the coherence between magnetic sublevels. The parametric scattering has been studied in the case of nondegenerate polarized waves.

Consider interaction of four elliptically polarized waves, that is two counter-propagating pump waves $E^{(1)}$ and $E^{(2)}$ and two counter-propagating signal waves $E^{(3)}$ and $E^{(4)}$ with a three-level resonant medium of which 1 is ground level with momentum J_1 and 2, 3 are doublet splitting excited levels with momenta J_2 and J_3 (for example, for sodium or potassium vapours $J_1 = 1/2$, $J_2 = 1/2$, $J_3 = 3/2$ and doublet splitting is of the order of 10 cm^{-1}). Pump waves are monochromatic with frequency ω_1 and signal waves are quasi-monochromatic with frequency spectrum spread near ω_1 , so that a spherical component of the electric field is

$$E_a(\vec{r}, t) = \frac{1}{2} \left[E_a^{(1)}(\vec{r}) e^{i\vec{k}_0 \vec{r}} + E_a^{(2)}(\vec{r}) e^{-i\vec{k}_0 \vec{r}} \right] e^{-i\omega_1 t}$$

$$+\frac{1}{2}\left[e^{i\vec{k}\vec{r}}\int E_{\alpha}^{(3)}(\omega_3,\vec{r})e^{i(\omega_1-\omega_3)t}d\omega_3+e^{-i\vec{k}\vec{r}}\int E_{\alpha}^{(4)}(\omega_4,\vec{r})e^{i(\omega_1-\omega_4)t}d\omega_4\right]e^{-i\omega_1 t}+\text{c.c.} \quad (1)$$

where: $\alpha = 0, \pm 1$, $E_0 = E_z$, $E_{\pm} = \pm(E_x \pm iE_y)/\sqrt{2}$.

The angle between the pump and signal directions of propagation is exceedingly large compared to the parametric interaction phase matching angle for each wave separately.

Solving the set of equations for the density matrix elements in the irreducible tensorial representation [6]–[8], we obtain the coefficient of the parametric coupling of the waves $K_2^{\omega_3}(q)$

$$K_2^{\omega_3}(q) = -\frac{g}{(2h)^3} \sum_{[\alpha,\beta,\gamma]} (-1)^{1-\alpha} C(11\kappa_1|q-\alpha q_1) C(11\kappa_1|\beta-\gamma q_1) \\ (E_{\alpha}^{(1)} E_{\beta}^{(2)} + E_{\alpha}^{(2)} E_{\beta}^{(1)}) E_{\gamma}^{(4)} (2\omega_1 - \omega_3) [|d_{21}|^4 a_{21} m_{21}^{\kappa_1} + |d_{31}|^4 a_{31} m_{31}^{\kappa_1} \\ + |d_{21}|^2 |d_{31}|^2 (\mu_{23}(\kappa_1) + \nu_{23}(\kappa_1))] \quad (2)$$

where:

$$a_{i1} = i \left[\frac{N_i}{2J_i+1} - \frac{N_i}{2J_i-1} \right], \quad i = 2, 3; \quad \alpha, \beta, \gamma = 0, \pm 1,$$

$$\mu_{23}(\kappa_1) = \begin{Bmatrix} 1 & 1 & \kappa_1 \\ j_1 & j_1 & j_3 \end{Bmatrix} \begin{Bmatrix} 1 & 1 & \kappa_1 \\ j_1 & j_1 & j_2 \end{Bmatrix} \\ \times \left[\frac{2\Gamma_i - \Gamma_{11}^{\kappa_1}}{(\Gamma_1 - i\varepsilon_1)(\Gamma_1 - \Gamma_{11}^{\kappa_1} - i\varepsilon_1)(\Gamma_1 - \Gamma_{11}^{\kappa_1} + i\varepsilon_1)} \times \frac{1}{i(\omega_3 - \omega_1) + \Gamma_{11}^{\kappa_1}} \right. \\ \left. + \frac{2\Gamma_2 - \Gamma_{11}^{\kappa_1}}{(\Gamma_2 - i\varepsilon_2)(\Gamma_2 - \Gamma_{11}^{\kappa_1} - i\varepsilon_2)(\Gamma_2 - \Gamma_{11}^{\kappa_1} + i\varepsilon_2)} \times \frac{1}{i(\omega_3 - \omega_1) + \Gamma_{11}^{\kappa_1}} \right. \\ \left. + \frac{i}{2\varepsilon_1(\Gamma_{11}^{\kappa_1} - \Gamma_1 - i\varepsilon_1)} \times \frac{1}{i(\omega_3 - \omega_1 + \varepsilon_1) + \Gamma_1} \right. \\ \left. + \frac{i}{2\varepsilon_2(\Gamma_{11}^{\kappa_1} - \Gamma_2 - i\varepsilon_2)} \times \frac{1}{i(\omega_3 - \omega_1 + \varepsilon_2) + \Gamma_2} \right. \\ \left. + \frac{i(\Gamma_1 + i\varepsilon_1)}{2\varepsilon_1(\Gamma_1 - i\varepsilon_1)(\Gamma_1 - \Gamma_{11}^{\kappa_1} - i\varepsilon_1)} \times \frac{1}{i(\omega_3 - \omega_1 - \varepsilon_1) + \Gamma_1} \right. \\ \left. + \frac{i(\Gamma_2 + i\varepsilon_1)}{2\varepsilon_2(\Gamma_2 - i\varepsilon_2)(\Gamma_2 - \Gamma_{11}^{\kappa_1} - i\varepsilon_2)} \times \frac{1}{i(\omega_3 - \omega_1 - \varepsilon_2) + \Gamma_2} \right], \quad (3)$$

$$\begin{aligned}
v_{23}(\kappa_1) = & \left\{ \begin{matrix} 1 & 1 & \kappa_1 \\ j_3 & j_2 & j_1 \end{matrix} \right\} \left\{ \begin{matrix} 1 & 1 & \kappa_1 \\ j_3 & j_2 & j_1 \end{matrix} \right\} \\
& \times \left\{ \frac{\Gamma_1 + \Gamma_2 - \Gamma_{32}^{\kappa_1}}{(\varepsilon_1 + i\Gamma_2)[\Omega + \varepsilon_1 + i(\Gamma_2 - \Gamma_{32}^{\kappa_1})]} \times \frac{1}{\Omega - \varepsilon_2 + i(\Gamma_1 - \Gamma_{32}^{\kappa_1})} \right. \\
& \times \frac{1}{\omega_3 - \omega_1 - \Omega + i\Gamma_{32}^{\kappa_1}} + \frac{\Gamma_2 + \Gamma_1 - \Gamma_{23}^{\kappa_1}}{(\varepsilon_2 + i\Gamma_1)[\varepsilon_2 - \Omega + i(\Gamma_1 - \Gamma_{23}^{\kappa_1})]} \\
& \left. \times \frac{1}{i(\Gamma_2 - \Gamma_{23}^{\kappa_1}) - \varepsilon_1 - \Omega} \times \frac{1}{\omega_3 - \omega_1 + \Omega + i\Gamma_{23}^{\kappa_1}} \right\}, \quad (4)
\end{aligned}$$

where: $\Gamma_1 = \Gamma_{12}^{(1)} = \Gamma_{21}^{(1)}$, $\Gamma_2 = \Gamma_{13}^{(1)} = \Gamma_{31}^{(1)}$.

Here, $\varepsilon_{i-1} = \omega_1 - \omega_{i1}$ are the resonance detunings between the pump wave frequency ω_1 and the frequency of the atomic transitions ω_{i1} ($i = 2, 3$), $\Omega = \omega_{31} - \omega_{21}$ is a value of doublet splitting, N_i are atom densities, Γ_{ik}^{κ} are the constants of the relaxations of the polarization momenta, $\kappa_1 = 0$ corresponds to the population decay, $\kappa_1 = 1$ corresponds to orientation decay, $\kappa_1 = 2$ corresponds to the alignment decay and q is a projection of momentum κ , ω_3 is the carrier frequency of the signal wave, $C(11|q - \alpha q_1)$ and $C(11|\beta - \gamma q_1)$ are the Clebsch-Gordan coefficients, d_{ik} are the dipole matrix elements.

If changes of $|E_a^{(1,2)}|^2$ in medium are substantial, all solving equation parameters are coordinate functions due to their dependence on pump wave intensity [5]. As it is very difficult to detect and count these changes, then as a rule the equations are being solved based on the assumption that pump wave intensities are constant. Thus in the solution (2) the spatial oscillation averaging of the pump field is carried out.

The coefficient $m_{21}^{\kappa_1}$ for two-level medium was obtained by the authors in paper [9] and $m_{31}^{\kappa_1}$ can be obtained from $m_{21}^{\kappa_1}$ by exchanging index 2 with 3. From the obtained expression (2) one can see that the parametric coupling coefficient has a complicated structure. It contains three types of terms, that is, the term m_{21} which is due to resonance with the transition 1-2, m_{31} which is due to resonance with transition 1-3, the interference terms μ_{23} and v_{23} which are due to both resonances 1-2 and 1-3. The most interesting are the third type terms, which are caused by mutual influence of both doublet split sublevels of the excited state in the field of wave.

There are two reasons for appearance of these terms, i.e., nonlinear population of the level 1 resulting from dipole-allowed transitions 1-2 and 1-3 (the term μ_{23}), and nondiagonal transition current 2-3 connected to doublet splitting of the excited state (the term v_{23}).

One can see that, similar to the scalar case, the latter has collision induced nature and vanishes at pure radiational relaxation. Indeed, v_{23} is proportional to the expression $\Gamma_1 + \Gamma_2 - \Gamma_{23}^{\kappa}$. Such a factor is included in the expression that describes processes which have been firstly suggested by Bloembergen *et al.* in four-wave mixing processes in sodium vapours [10]. These processes are not due to real population of levels and occur as a result of coherence of density matrix nondiagonal element.

Let us focus on spectral pattern of the reflected wave.

Similar to the scalar case, a set of resonant poles appear which correspond to the following processes of interaction of the waves with the atom, namely, absorption of two counter-propagating pump quanta with frequency ω_1 , and radiation of: 1) two quanta with the same frequency, 2) a quantum with frequency close to atomic absorption frequency 1–2 and a quantum with frequency close to three-photon scattering frequency of the same transition, 3) a quantum with frequency close to atomic absorption frequency 1–3 and a quantum with frequency close to three-photon scattering frequency of the same transition, and 4) two quanta with frequencies shifted by a magnitude of doublet splitting with respect to the pump frequency, which are due to the collision induced scattering process.

It is to be mentioned that three of these poles corresponding to Rayleigh and collision induced scattering processes are split by magnitudes of $\Gamma_{11}^{(\times)}$ and $\Gamma_{23}^{(\times)}$, $\Gamma_{32}^{(\times)}$ correspondingly, depending on wave polarization. This fact can be used for determination of relaxation constants of polarization momenta of dipole-forbidden atomic transition 2–3: Γ_{23}^{\times} , Γ_{32}^{\times} . This is a very complicated problem in atomic and molecular spectroscopy.

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