The influence of polarizability on saturated absorption at forbidden atomic transitions

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The theory for saturated magnetic dipole and electric quadrupole absorption of monochromatic radiation at dipole-forbidden transitions of gas media is developed. It is shown that in the case of quasi-resonant polarizability, when the radiation frequency is close to the frequency of the transition from the upper level of a forbidden transition to the next upper state, the line shift for the absorption at dipole-forbidden transition can reach a large magnitude. The possibility of radiation amplification without population inversion at dipole-forbidden and quasi-resonant transitions is predicted for sufficiently high intensities and small frequency detuning from the eigenfrequency of quasi-resonant transition.

Introduction

Magnetic dipole and electric quadrupole absorption at forbidden electric dipole transitions in atomic gases and iron vapors can be quite noticeable and saturable at small intensities of resonance radiation in case of small atom densities. The usual reason is long lifetimes of forbidden transition levels, which can be three order of magnitude longer than lifetimes of dipole-allowed transition levels.[1] This compensates a small value of radiation interaction with forbidden transitions. The values of atomic magnetic dipole and electric quadrupole moments can be determined on the base of quantitative measurements of absorption at forbidden transitions. Radiation, resonant to a forbidden transition, can be also used as a closed field in problems of nonlinear optics and nonlinear spectroscopy, based on ring frequency mixing.[2,3] Assess the order of magnitude of saturation intensity I_{sat} from the condition $\kappa = 1$ (κ is the saturation parameter) considering the saturation as magnetic dipole:

$$\kappa \equiv \frac{I}{I_{\text{sat}}} = \frac{1}{\Gamma\gamma}\left(\frac{mB}{\hbar}\right)^2 = 1, \quad I_{\text{sat}} = \frac{c\hbar^2\Gamma\gamma}{8\pi m^2}, \qquad (1)$$

where I is the radiation intensity; B is the light wave magnetic induction; γ and Γ are the longitudinal and transversal relaxation constants of the forbidden transition; m is the matrix element of the magnetic dipole moment operator; c is the light speed; $\hbar$ is the Planck constant. Taking $m \sim \mu_B \sim 10^{-20}$ CGSE (μ_B is the Bohr magneton) and $\gamma \sim \Gamma \sim 10^7$ s^{-1}, we obtain $I_{\text{sat}} \sim 1$ W/cm^2 from Eq. (1). Pulsed lasers, used in spectroscopy of gaseous media, are often have radiation intensity of about 1 MW/cm^2 and higher.

Thus, nonlinear spectroscopy of forbidden atom transitions is quite possible. In view of this, there appears a necessity of proper description of saturated absorption at forbidden transitions on the base of quantum kinetic equations for the medium statistical matrix, commonly used in nonlinear spectroscopy. Such description is absent in the available literature. A probable reason is nontrivial character of radiation–forbidden transition interaction, where electric and magnetic light wave components participate simultaneously. Field vector potential and current density are widely used in this case. However, such description for nonresonance interactions, e.g., in case of stimulated Raman scattering, produces results essentially differing from those obtained from the description based on observable electric field strength, magnetic induction, as well as magnetic dipole and electric quadrupole atom moments.[4–6] Physical nature of this difference is still unknown.

Besides, multipole atom–radiation field interaction is considered in the literature[7,8] only for the case of spherical waves and is expressed in terms of vector potential. Such representation is good for application to calculations of radiation lifetime of atom levels.[9] For most laser spectroscopy problems, the Hamiltonian of atom interaction with plane waves is required. The Hamiltonian for a plane classical wave, written in the complex form, has been derived in Ref. 10. However, to describe nonresonant interactions in cases of inapplicability of rotating-wave approximation (RWA), the field is required to be presented in the real form. The corresponding description for gaseous media is derived in Section 1 of this work.

Since pulsed laser radiation of sufficiently high intensity is supposed to be used in the study of saturated absorption at forbidden transitions, it is necessary to know, in what way and how strong this absorption is influenced by nonresonance allowed transitions to the level 3, adjacent with the

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considered one 1–2, i.e., atom polarizability. This influence can be especially noticeable in quasi-resonance conditions:

$$\omega_{31} - 2\omega \approx \omega_{32} - \omega \sim \Delta,$$

where $\omega \approx \omega_{21}$ is the radiation frequency; ω_{21} is the forbidden transition frequency; ω_{31} and ω_{32} are the frequencies of nonresonance allowed transitions; Δ is some critical frequency, detuning which is to be determined (see the Figure 1).

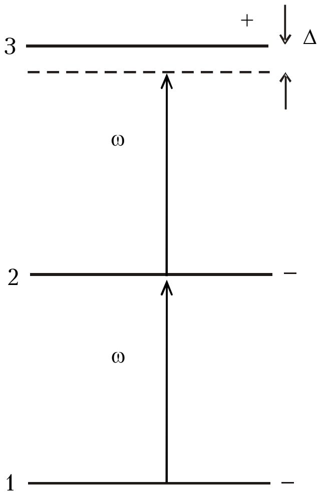

Fig. 1. Scheme of ω–radiation interaction with the resonance dipole-forbidden transition 1–2 and nonresonance allowed transition 2–3. Parity of levels 1, 2, and 3 is conventionally marked by «+» and «–».

Relatively small magnitude Δ can exist for both transitions from the ground state to the first metastable level 2 (e.g., for copper atom[11]) and for “hot” forbidden transitions from upper states in gas plasma. The value of field interaction with dipole-allowed transitions at large radiation intensities is much greater than with the forbidden transition, therefore, Δ detuning overcoming is quite possible. As a consequence, the forbidden transition stops to be an isolated system, which should be taken into account when comparing the theory with experiments. The character and extent of polarizability influence on absorption at forbidden transition are determined in Section 2 on the base of the corresponding theory.

1. Magnetic dipole and electric quadrupole interactions of a plane light wave with the forbidden transition

Derive the Hamiltonian of field interaction with the forbidden transition by the commonly used scheme,[7,8,10] taking the Lorentz force as a basis,[12] which includes the observable light-wave electric field strength $\mathscr{E}$ and magnetic induction $\mathscr{B}$:

$$\mathbf{F} = e\mathscr{E} + \frac{e}{c}[\mathbf{v}\mathscr{B}], \qquad (2)$$

where e and $\mathbf{v}$ are the charge and the speed of an electron in the atom; square brackets mean the vector product. Write vectors $\mathscr{E}$ and $\mathscr{B}$ as

$$\begin{gathered}\mathscr{E}(\mathbf{r},t) = \mathbf{E}(\xi)\cos\Psi, \quad \mathscr{B}(\mathbf{r},t) = \mathbf{B}(\xi)\cos\Psi; \\ \Psi \equiv \omega t - \mathbf{kr}, \quad \xi \equiv \mathbf{nr} - \omega t/k, \quad \mathbf{n} = \mathbf{k}/k,\end{gathered} \qquad (3)$$

where $\mathbf{k}$ is the wave vector of the wave.

Present the radius vector $\mathbf{r}$ as

$$\mathbf{r} = \mathbf{r}_0 - \mathbf{r}_1, \quad |\mathbf{r}_1| \approx a_0, \qquad (4)$$

where $\mathbf{r}_0$ is the distance to the atom center; $\mathbf{r}_1$ presets the electron position in the atom; a_0 is the Bohr radius.

An interaction potential is determined through the corresponding transformation and analysis of the matrix element of interaction energy of electrons in the atom with the light wave field $U = -\,\mathbf{r}_1\mathbf{F}$ [Refs. 7, 8, 10, and 12] (omit the assumed sum over electrons). As the Lorentz force is basic in the description, required transformations reduce to expansion of the phase Ψ and amplitude $\mathbf{E}$ of the electric field in terms of small $\mathbf{r}_1$ with keeping only first degrees of smallness. Such expansion is not required for the magnetic field, since the ratio of the second item in Eq. (2) to the first one is small: of order of the fine-structure constant. Applying Eqs. (2)–(4), the matrix element of interaction potential is

$$\begin{aligned}U_{mn} &= -e\int \Psi_m^* \left\{ \mathbf{r}_1 \left[\mathscr{E}_0 - (\mathbf{kr}_1)\mathbf{E}(\xi_0)\sin\Psi_0 + \right.\right. \\ &\left.\left. + \frac{1}{k}\frac{\mathrm{d}\mathbf{E}(\xi_0)}{\mathrm{d}\xi_0}\cos\Psi_0 \right] + \frac{1}{c}[\mathbf{r}_1\mathbf{v}]\mathscr{B}_0 \right\} \Psi_n \mathrm{d}\mathbf{r}_1 = \\ &= -e\int \Psi_m^* \left[\mathbf{r}_1\mathscr{E}_0 - \mathbf{r}_1(\mathbf{r}_1\nabla)\mathscr{E}_0 + \frac{1}{c}[\mathbf{r}_1\mathbf{v}]\mathscr{B}_0 \right] \Psi_n \mathrm{d}\mathbf{r}_1. \qquad (5)\end{aligned}$$

The subscript «0» means $\mathbf{r} = \mathbf{r}_0$ for the corresponding variables. The differential term in Eq. (5) corresponds to the electric quadrupole interaction.

According to the definitions of electric and magnetic dipole moments and electric quadrupole moment,[10,12] the operators

$$\begin{gathered}e\mathbf{r}_1 \to \hat{\mathbf{d}}, \quad e\mathbf{r}_1(\mathbf{r}_1\nabla) \to \frac{1}{6}(\hat{\mathbf{Q}}\nabla), \\ e\mathbf{v} \to \hat{\mathbf{j}}, \quad \frac{1}{c}[\mathbf{r}_1\hat{\mathbf{j}}] = \hat{\mathbf{m}} \to \hat{\mathbf{m}} + \hat{\mathbf{s}}\end{gathered} \qquad (6)$$

correspond to three items in integral (5).

In (6) $\hat{\mathbf{d}}$ and $\hat{\mathbf{Q}}$ are the operators of electric dipole and quadrupole moments; $\hat{\mathbf{j}}$ is the operator of current density; $\hat{\mathbf{m}}$ is the operator of orbital magnetic dipole moment, which should be added by the spin $\hat{\mathbf{s}}$. Hence, the operator of plane wave–atom interaction has the form

$$\hat{U} = -\hat{\mathbf{d}}\mathscr{E} - \hat{\mathbf{m}}\mathscr{B} + \frac{1}{6}(\nabla\hat{\mathbf{Q}})\mathscr{E}. \qquad (7)$$

If change $\nabla \to i\mathbf{k}$ when using complex field presentation and resonance RWA, equation (7) coincides with the equation for interaction operation, given in Ref. 10.

Note, that the choice rules in calculations of matrix elements of magnetic dipole and electric quadrupole moment operators differ in general case,[13] but matrix elements of both these moments are simultaneously nonzero for many dipole-forbidden transitions. Just such situation is considered below.

Consider now the wave equations. Consider two first Maxwell equations, describing waves in absorbing media without extraneous charges[14,15]:

$$\begin{gathered}\mathrm{rot}\mathscr{B} - \frac{1}{c}\frac{\partial\mathscr{E}}{\partial t} = \frac{4\pi}{c}\frac{\partial\mathbf{P}}{\partial t} + 4\pi\,\mathrm{rot}\,\mathbf{M},\\ \mathrm{rot}\mathscr{E} + \frac{1}{c}\frac{\partial\mathscr{B}}{\partial t} = 0.\end{gathered} \qquad (8)$$

Here $\mathbf{P}$ is the polarization and $\mathbf{M}$ is the medium magnetization.

For definiteness, consider linearly polarized radiation. As it is common for problems with medium quantization, take z axis parallel to the direction of electric field amplitude $\mathscr{E}$. In this case, the wave vector $\mathbf{k}$ is y-axially directed and the magnetic induction $\mathscr{B}$ is x-axially directed. Hence, the vectors of electric and magnetic light wave components are represented as

$$\begin{gathered}\mathscr{E} = \big(0, 0, E(y)\cos\Psi\big);\\ \mathscr{B} = \big(B, 0, 0\big), \quad B = B_\mathrm{c}(y)\cos\Psi + B_\mathrm{s}(y)\sin\Psi,\\ \Psi = \omega t - ky.\end{gathered} \qquad (9)$$

Applying second equations (8) and (9), obtain an exact expression for magnetic induction in terms of the electric field amplitude:

$$\begin{gathered}B = \frac{c}{\omega}\left(kE\cos\Psi - \frac{\mathrm{d}E}{\mathrm{d}y}\sin\Psi\right),\\ B_\mathrm{c} = \frac{ck}{\omega}E, \quad B_\mathrm{s} = -\frac{c}{\omega}\frac{\mathrm{d}E}{\mathrm{d}y}.\end{gathered} \qquad (10)$$

Similar to Eq. (9), the polarization and magnetization are defined as

$$\begin{gathered}\mathbf{P} = \big(0, 0, P\big), \quad P = P_\mathrm{c}\cos\Psi + P_\mathrm{s}\sin\Psi;\\ \mathbf{M} = \big(M, 0, 0\big), \quad M = M_\mathrm{c}\cos\Psi + M_\mathrm{s}\sin\Psi.\end{gathered} \qquad (11)$$

Substituting Eqs. (9)–(11) in Eq. (8), neglecting the second derivatives of slowly varying coordinate amplitudes, and equaling the coefficients sinΨ and cosΨ to zero (which is equivalent to averaging over fast phase oscillations), obtain two equations:

$$\begin{gathered}2k\frac{\mathrm{d}E}{\mathrm{d}y} = -\frac{4\pi\omega^2}{c^2}\left(P_\mathrm{s} + \frac{kc}{\omega}M_\mathrm{s} - \frac{c}{\omega}\frac{\mathrm{d}M_\mathrm{c}}{\mathrm{d}y}\right);\\ \left(k^2 - \frac{\omega^2}{c^2}\right)E = \frac{4\pi\omega^2}{c^2}\left(P_\mathrm{c} + \frac{kc}{\omega}M_\mathrm{c} + \frac{c}{\omega}\frac{\mathrm{d}M_\mathrm{s}}{\mathrm{d}y}\right).\end{gathered} \qquad (12)$$

The first equation in (12) is the wave equation for slowly varying electric field amplitude, and the second one is the dispersion equation, from which the frequency dependence of wave number or refraction index $n_d \equiv kc/\omega$ is determined. The refraction index n_d in absorption at allowed transitions in gases at the atmospheric pressure differs from unit insignificantly and is equal to 10^{-3}–10^{-4} [Ref. 16]. It is 4–6 orders of magnitude less at forbidden transitions, therefore, $n_d = 1$ with good accuracy. Thus, neglecting the dispersion when absorbing at forbidden transitions, $k = \omega/c$. Corrections in Eqs. (10) and (12) due to coordinate derivative of amplitudes are of the same small magnitude, since these derivatives enter the equations with the coefficient $\lambda\!\!\!^{-} = c/\omega$.

Assuming amplitude variations small at sizes of the order of wavelength $\lambda\!\!\!^{-}$ and omitting these derivatives, we obtain a tight correlation between the light wave magnetic and electric components:

$$B_\mathrm{c} \cong E, \quad B_\mathrm{s} \cong 0, \quad B \cong E\cos\Psi. \qquad (13)$$

Due to these correlation, it is sufficient to solve only first equation (12) for electric field amplitude to determine the absorption at the forbidden transition; the equation for the radiation intensity I remains the same: $I = cE^2/(8\pi)$. The performed approximations result in a simple form of first equation (12), which agrees with the form of wave equation for slowly varying amplitude in case of absorption at dipole-allowed transitions. The only difference is in the right part of Eq. (12), where the polarization P_s, related to sine, is changed to the sum of this polarization with the magnetization M_s:

$$\begin{gathered}2\frac{\mathrm{d}E}{\mathrm{d}y} = -\frac{4\pi\omega}{c}\mathsf{P}_\mathrm{s},\\ \mathsf{P}_\mathrm{s} \equiv P_\mathrm{s} + M_\mathrm{s}.\end{gathered} \qquad (14)$$

The magnitude $\mathsf{P} = \mathsf{P}_\mathrm{s}\sin\Psi + \mathsf{P}_\mathrm{c}\cos\Psi$ is defined as the quantum mean of the effective dipole moment operator $\hat{D}$, which can be determined on the base of interaction Hamiltonian (7) with accounting for Eq. (13), which will be done after analysis of equations for statistic matrix of two-level quantum system.

The quantum system in the considered case consists of the lower and upper levels 1 and 2, transition between which is forbidden for electric dipole interaction and is allowed for magnetic dipole and electric quadrupole interactions. A nonzero off-diagonal matrix element of interaction Hamiltonian (7) can be written in the form

$$-U_{12} = m_{12}\mathscr{B} - \frac{1}{6}Q_{12}\nabla\mathscr{E} = E\left(m_{12} - \frac{1}{6}Q_{12}\nabla\right)\cos(\omega t - ky) =$$
$$= E\left[m_{12}\cos(\omega t - ky) - \frac{1}{6}kQ_{12}\sin(\omega t - ky)\right] =$$
$$= E(m\cos\Psi - q\sin\Psi) \equiv U,$$
$$m \equiv m_{12}, \quad q \equiv \frac{1}{6}kQ_{12}. \qquad (15)$$

In accordance with the made approximations, the coordinate derivative of slowly varied amplitude is not taken into account here, and the gradient ∇ acts only on the phase Ψ.

The statistical matrix equation for the two-level system in the model of relaxation constants for homogeneous broadening has the form

$$\begin{cases} \dot{n} + \gamma n = \gamma n_0 + \dfrac{4}{\hbar}U\,\mathrm{Re}\,i\rho_{21}, \\ \dot{\rho}_{21} + (\Gamma + i\omega_{21})\rho_{21} = \dfrac{i}{\hbar}Un, \end{cases} \qquad (16)$$
$$n \equiv \rho_1 - \rho_2.$$

Here n is the population difference between levels 1 and 2; n_0 is the equilibrium population difference; ρ_{21} is the off-diagonal element of statistical matrix, or forbidden transition polarization up to a constant multiplier.

Pass from Eq. (16) to time independent equations by means of isolation of fast polarization oscillations of the ρ_{21} transition:

$$\rho_{21} = r\,\mathrm{e}^{-i\Psi}, \quad r = r' + ir''. \qquad (17)$$

After substituting Eq. (17) in Eq. (16) and applying RWA, obtain

$$\begin{cases} n + \dfrac{2E}{\hbar\gamma}(mr'' + qr') = n_0, \\ (\Gamma - i\delta)r = \dfrac{iE}{2\hbar}(m - iq)n, \end{cases} \qquad (18)$$
$$\delta \equiv \omega - \omega_{21}.$$

Compare Eq. (18) with the similar time independent equations for the two-level system in case of dipole-allowed 1–2 transition:

$$\begin{cases} n - \dfrac{2E}{\hbar\gamma}\mathrm{Re}(id_{12}r) = n_0, \\ (\Gamma - i\delta)r = \dfrac{iE}{2\hbar}d_{21}n. \end{cases} \qquad (19)$$

As it follows from the comparison of Eqs. (18) and (19), the forms of these sets of equations are the same, if matrix elements of the effective dipole moment operator $\hat{D}$ are defined as complex variables

$$D_{21} = m - iq, \quad D_{12} = m + iq. \qquad (20)$$

Actually,

$$-\mathrm{Re}(iD_{12}r) = -\mathrm{Re}\left[i(m + iq)(r' + ir'')\right] = mr'' + qr' \quad (21)$$

follows from Eq. (20) for $\mathrm{Re}(iD_{12}r)$, proportional to the work of the field[17] or P_s, and agrees with the expression in parenthesis in the first equation (18). Correspondingly, as applied to absorption at forbidden transitions, the quantum mean of the effective dipole moment operator or generalized macroscopic medium polarization can be written as

$$P = N\,Sp\hat{D}\hat{\rho} = N(D_{12}\rho_{21} + D_{21}\rho_{12}) = 2N\,\mathrm{Re}(D_{12}\rho_{21}) =$$
$$= 2N\,\mathrm{Re}\left[(m + iq)(r' + ir'')(\cos\Psi - i\sin\Psi)\right] =$$
$$= 2N\left[(mr' - qr'')\cos\Psi + (mr'' + qr')\sin\Psi\right]; \qquad (22)$$
$$P_s = 2N(mr'' + qr').$$

Here N is the density of radiation-interacting atoms.

The solution of Eq. (18) is

$$n = n_0\frac{\Gamma^2 + \delta^2}{\mathrm{Det}}; \quad r' = \frac{n_0E}{2\hbar}\frac{\Gamma q - \delta m}{\mathrm{Det}}; \quad r'' = \frac{n_0E}{2\hbar}\frac{\Gamma m + \delta q}{\mathrm{Det}};$$
$$\mathrm{Det} = \Gamma^2(1 + \kappa) + \delta^2, \quad \kappa = \frac{(m^2 + q^2)E^2}{\hbar^2\Gamma\gamma}. \qquad (23)$$

As is evident from Eq. (23), the saturation parameter κ is similar to those in case of absorption at an allowed transition, except for the squared dipole moment, which is changed here to the squared module of the matrix element of effective dipole moment (20), $|D_{12}|^2 = m^2 + q^2$. Substitution of Eqs. (22) and (23) in Eq. (14) results in the equation for the saturated absorption coefficient at a dipole-forbidden transition

$$\alpha = \frac{4\pi\omega}{c}\frac{P_s}{E} = \frac{4\pi Nn_0\omega\Gamma\left(m^2 + q^2\right)}{\Gamma^2(1 + \kappa) + \delta^2}. \qquad (24)$$

Equation (24) coincides with the Karplus–Schwinger formula for the coefficient of saturated absorption at an allowed transition[18] when changing $d^2 \to m^2 + q^2$ in the latter. Note, that $\alpha \propto (m^2 + q^2)/\Gamma$ in case of linear absorption in the line center from Eq. (24), while $\alpha \propto d^2/\Gamma_a$ (Γ_a is the transversal constant of the allowed transition relaxation) for a dipole-allowed transition. The ratio $(m^2 + q^2)/d^{\,2} \sim 10^{-4}$ while $\Gamma/\Gamma_a \le 10^{-2}$. Hence, the linear absorption coefficient at a forbidden transition is 2–3 orders of magnitude lower than those at an allowed transition. However, the ratio of the corresponding saturation parameters

$$\kappa/\kappa_a \propto [(m^2 + q^2)/d^{\,2}](\Gamma_a\gamma_a/\Gamma\gamma)$$

at $\gamma_a \sim \Gamma_a$, $\gamma \sim \Gamma$, and $\Gamma/\Gamma_a \le 10^{-2}$ can be about unit. The same follows from the assessments made in Introduction on the base of Eq. (1). In other words, absorption at forbidden transitions is much weaker than at allowed ones, but its saturation is reached at the same or a little larger intensity values.

As is evident from the analysis of absorption at forbidden transitions, a general scheme and results of saturated absorption coefficient calculation remain the same as for absorption at dipole-allowed transitions, despite the difference in the radiation–atom interaction mechanisms. This similarity could be revealed after expressing magnetic induction in terms of electric field strength (13) and introducing generalized medium polarization $P \to P + M \equiv P$ [Eqs. (14), (22)], and generalized dipole transition moment $d \to D$ [Eq. (20)]. Let us perform some transformations of the introduced variables to reduce statistical matrix equation (18) to a more compact form, completely coinciding with Eq. (19): write the matrix element of interaction Hamiltonian (15) as

$$U = E(m\cos\Psi - q\sin\Psi) = \sqrt{m^2 + q^2}E \times$$

$$\times\left(\frac{m}{\sqrt{m^2+q^2}}\cos\Psi - \frac{q}{\sqrt{m^2+q^2}}\sin\Psi\right) =$$

$$= \sqrt{m^2+q^2}E(\cos\chi\cos\Psi - \sin\chi\sin\Psi); \qquad (25)$$

$$U = \mu E\cos(\Psi + \chi),$$

$$\mu \equiv \sqrt{m^2+q^2}, \quad \chi \equiv \arctan\frac{q}{m}.$$

This form makes evident the fact that quadrupole interaction results in the χ-shift of polarization wave phase. This is due to differential character of quadrupole interaction operator [Eqs. (7) and (15)] and results in complex matrix elements of effective dipole moment operator (20) in conditions of resonance interaction (RWA applicability). Maximum phase shift χ is equal to $\pi/2$ at zero magnetic dipole moment.

Write matrix elements of effective dipole moment (20) in a similar form:

$$D_{12} = m + iq = \sqrt{m^2+q^2}\left(\frac{m}{\sqrt{m^2+q^2}} + \frac{iq}{\sqrt{m^2+q^2}}\right) =$$

$$= \mu(\cos\chi + i\sin\chi) = \mu e^{i\chi}, \qquad (26)$$

$$D_{21} = \mu e^{-i\chi}.$$

Again, to pass from time dependent equations for statistical matrix (16) to time independent ones instead of Eq. (17), assume

$$\rho_{21} = re^{-i(\Psi+\chi)}, \qquad (27)$$

and instead of Eq. (18) obtain time independent equations, completely coinciding with Eq. (19) for dipole-allowed transitions,

$$\begin{cases} n + \dfrac{2\mu E}{\hbar\gamma}r'' = n_0, \\ (\Gamma - i\delta)r = \dfrac{i\mu E}{2\hbar} \end{cases} \qquad (28)$$

with the solution

$$n = n_0\frac{\Gamma^2 + \delta^2}{\mathrm{Det}}; \quad r' = n_0\frac{\mu E}{2\hbar}\frac{\delta}{\mathrm{Det}}, \quad r'' = n_0\frac{\mu E}{2\hbar}\frac{\Gamma}{\mathrm{Det}},$$
$$\mathrm{Det} = \Gamma^2(1 + \kappa) + \delta^2, \quad \kappa = \frac{\mu^2E^2}{\hbar^2\Gamma\gamma}. \qquad (29)$$

As a result, polarization equation (22) is also simplified:

$$P = N(D_{12}\rho_{21} + D_{21}\rho_{12}) =$$

$$= 2N\,\mathrm{Re}(D_{12}\rho_{21}) = 2N\,\mathrm{Re}\left[\mu e^{i\chi}r e^{-i(\Psi+\chi)}\right] =$$

$$= 2N\mu\left[(r' + ir'')(\cos\Psi - i\sin\Psi)\right] =$$

$$= 2N\mu(r'\cos\Psi + r''\sin\Psi), \qquad (30)$$

$$P_s = 2N\mu r''.$$

Substitution of Eqs. (29) and (30) in absorption coefficient (24) with accounting for the expression for μ in Eq. (25) naturally gives the same result.

2. Polarizability influence

Let us define the character and the extent of polarizability influence on the coefficient of saturated radiation absorption at the forbidden transition (24) on the base of three-level atom model (see Figure). The equations for statistical matrix of the three-level Λ-system were used in Ref. 2 to describe stimulated Raman two-wavelength scattering, closed by radiation, interacting with a resonance forbidden transition through the magnetic dipole mechanism. Generalizing these equations to the case of simultaneous interaction of the resonance field with atom magnetic dipole and electric quadrupole moments on the base of the results from Section 1, obtain

$$\begin{cases} \dot\rho_1 + \gamma(\rho_1 - \rho_1^0) = \dfrac{2}{\hbar}(d_1\mathscr{E}\,\mathrm{Re}\,i\rho_{31} + U\,\mathrm{Re}\,i\rho_{21}), \\ \dot\rho_2 + \gamma(\rho_2 - \rho_2^0) = \dfrac{2}{\hbar}(d_2\mathscr{E}\,\mathrm{Re}\,i\rho_{32} - U\,\mathrm{Re}\,i\rho_{21}), \\ \rho_1 + \rho_2 + \rho_3 = 1, \\ -i\dot\rho_{31} + \omega_{31}\rho_{31} = \dfrac{1}{\hbar}\left[\mathscr{E}(d_1n_{13} + d_2\rho_{21}) - U\rho_{32}\right], \\ -i\dot\rho_{32} + \omega_{32}\rho_{32} = \dfrac{1}{\hbar}\left[\mathscr{E}(d_2n_{23} + d_1\rho_{21}^*) - U\rho_{31}\right], \\ \dot\rho_{21} + (\Gamma + i\omega_{21})\rho_{21} = \dfrac{i}{\hbar}\left[Un_{12} + \mathscr{E}(d_2\rho_{31} - d_1\rho_{32}^*)\right], \end{cases} \qquad (31)$$

$$n_{12} = \rho_1 - \rho_2, \quad n_{13} = \rho_1 - \rho_3 = 2\rho_1 + \rho_2 - 1,$$

$$n_{23} = \rho_2 - \rho_3 = \rho_1 + 2\rho_2 - 1,$$

$$\mathscr{E} = E\cos\Psi, \quad U = \mu E\cos(\Psi + \chi), \quad \Psi = \omega t - ky.$$

Here ρ_j, $j = 1, 2, 3$ are the level populations; ρ_j^0 are the equilibrium level populations in the absence of radiation; ρ_{31} and ρ_{32} are the polarizations of nonresonance allowed transitions; ρ_{21} is the forbidden transition polarization; d_1 and d_2 are the matrix elements of the operators of electric dipole moments at transitions 1–3 and 2–3, respectively; ω_{ij} and n_{ij} are the frequencies and population level differences for the transitions i–j; U, μ, and χ are defined by Eq. (25); Γ is the transversal relaxation constant of forbidden transition polarization. Obtain the time independent equations from Eq. (31) applying RWA and isolating fast polarization oscillations:

$$\rho_{31} = R_1 \exp(-2i\Psi), \quad \rho_{32} = R_2 \exp(-i\Psi),$$
$$\rho_{21} = r\exp\left[-i(\Psi + \chi)\right]. \tag{32}$$

Substitution of Eq. (32) in Eq. (31) results in

$$\begin{cases} \rho_1 + \dfrac{\mu E}{\hbar\gamma} r'' = \rho_1^0, \\[2mm] \rho_2 + \dfrac{d_2 E}{\hbar\gamma} R_2'' - \dfrac{\mu E}{\hbar\gamma} r'' = \rho_2^0, \\[2mm] R_1 = \dfrac{E}{2\hbar\Delta} \mathrm{e}^{-i\chi} (d_2 r - \mu R_2), \\[2mm] R_2 = \dfrac{E}{2\hbar\Delta}\left(d_2 n_{23} - \mu R_1 \mathrm{e}^{i\chi}\right), \\[2mm] (\Gamma - i\delta) r = \dfrac{iE}{2\hbar}\left(\mu n_{12} + d_2 R_1 \mathrm{e}^{i\chi}\right), \end{cases} \tag{33}$$

$$\Delta \equiv \omega_{31} - 2\omega \cong \omega_{32} - \omega, \quad \delta \equiv \omega - \omega_{21}.$$

A target quantity is the part P_s of generalized polarization P (14), determining the absorption coefficient and having the form

$$P = 2N \operatorname{Re}\left(d_1\rho_{31} + d_2\rho_{32} + \mu \mathrm{e}^{i\chi}\rho_{21}\right) =$$
$$= 2N\left[(d_2 R_2' + \mu r')\cos\Psi + (d_2 R_2'' + \mu r'')\sin\Psi\right]; \tag{34}$$
$$P_\mathrm{s} = 2N(\mu r'' + d_2 R_2'')$$

in the considered case of three-level atom; here 2ω-oscillating terms are omitted.

We have

$$R_1 = \frac{d_2 E}{2\hbar\Delta} \mathrm{e}^{-i\chi}\left(r - \frac{\mu E}{2\hbar\Delta} n_{23}\right),$$
$$R_2 = \frac{d_2 E}{2\hbar\Delta}\left(n_{23} - \frac{\mu E}{2\hbar\Delta} r\right) \tag{35}$$

from the second and third equations (33) in the approximation $\kappa_\mu \equiv \left(\dfrac{\mu E}{2\hbar\Delta}\right)^2 \ll 1$, which is quite satisfactory ($\kappa_\mu \sim 10^{-2}$–10^{-8}) at a radiation intensity of 100 MW/cm^2, $\mu \sim 10^{-20}$ CGSE, and Δ between 1 and 10^3 cm^{-1}. Substitution of Eq. (35) in Eq. (33) and transition from level populations to population differences according to Eq. (31) result in the following equations:

$$\begin{cases} n_{12} + \xi V(2 + w) r'' = n_{12}^0, \\ n_{23} - \xi V(1 + 2w) r'' = n_{23}^0, \\ (1 - i\delta_1) r = iV(n_{12} - w n_{23}), \end{cases} \tag{36}$$

$$V \equiv \frac{\mu E}{2\hbar\Gamma}, \quad w \equiv \left(\frac{d_2 E}{2\hbar\Delta}\right)^2, \quad \xi \equiv \frac{2\Gamma}{\gamma}, \quad \delta_1 \equiv \frac{\delta + \Delta w}{\Gamma}.$$

Equations (36) coincide with Eqs. (28) for the isolated transition 1–2 to within notation at $w \to 0$. The solution of Eq. (36) is

$$n_{12} = \frac{1}{\mathrm{Den}}\left\{\left[1 + w(1+2w)W\right] n_{12}^0 + w(2+w) W n_{23}^0\right\},$$
$$n_{23} = \frac{1}{\mathrm{Den}}\left\{(1+2w) W n_{12}^0 + \left[1 + (2+w)W\right] n_{23}^0\right\},$$
$$r = r' + ir'' = \frac{V}{1+\delta_1^2} \frac{-\delta_1 + i}{\mathrm{Den}}\left(n_{12}^0 - w n_{23}^0\right), \tag{37}$$
$$\mathrm{Den} \equiv 1 + 2\left(1 + w + w^2\right) W, \quad W \equiv \frac{\xi V^2}{1+\delta_1^2}.$$

From Eq. (35) we have

$$R_2'' = -\frac{\mu d_2 E^2}{4\hbar^2\Delta^2} r'' = -w \frac{\mu}{d_2} r''. \tag{38}$$

Substitution of Eqs. (37), (38), and (34) in the absorption coefficient expression

$$\alpha = \frac{4\pi\omega}{c} \frac{P_\mathrm{s}}{E}, \tag{39}$$

following from shortened wave equations (14), gives the final equation for the absorption coefficient:

$$\alpha = \alpha_0 \frac{(1 - w)\left(n_{12}^0 - w n_{23}^0\right)\Gamma^2}{\Gamma^2\left[1 + 2\xi\left(1 + w + w^2\right)V^2\right] + (\delta + w\Delta)^2},$$
$$\alpha_0 = \frac{4\pi N \mu^2 \omega}{c\hbar\Gamma}. \tag{40}$$

At $w \to 0$ Equation (40) coincides with Eq. (24) and, hence, the influence of atom polarizability on saturated absorption at forbidden transitions is completely characterized by the only parameter w. Estimates of the value of saturation parameter w (36) at forbidden transitions at $I = 100$ MW/cm^2, $d_2 = 1$ D, and frequency detuning $\Delta = 10$ cm^{-1}, give $w \approx 0.05$; the parameter $w \propto 1/\Delta^2$ is larger than unit ($w \approx 5$) at smaller $\Delta = 1$ cm^{-1}. The coefficient $1 - w$ in numerator (40) alters its sign in this case, and the absorption coefficient α becomes negative (for the transitions 1–2 and 2–3, the equilibrium population differences is considered as corresponding to the Boltzmann distribution and positive, like the difference $n_{12}^0 - w n_{23}^0$). In other words, the radiation amplification instead of absorption at the dipole-forbidden transition is probable in the conditions of polarizability quasi-resonance for sufficiently high radiation intensities of about 100 MW/cm^2 and small frequency detuning $|\Delta| = |2\omega - \omega_{31}| \approx |\omega - \omega_{32}| \sim 1$ cm^{-1}.

The coefficient of amplification at $w \sim 2$ is determined by small α_0 (40) and is to be small as well. For example, $\alpha \sim 2 \cdot 10^{-6}$ cm^{-1} when $N \sim 10^{17}$ cm^{-3}, $\mu \sim 10^{-20}$ CGSE, $\Gamma \sim 0.6 \cdot 10^9$ s^{-1}, and $\lambda \sim 0.5 \cdot 10^{-4}$ cm.

Of more interest is the probability of radiation amplification without inversion of populations due to interference of polarizations of forbidden (r'') and quasi-resonance allowed (R_2'') transitions, which follows from Eq. (34) for the medium polarization P_s. In contrast to nonlinear interference of Λ-atom polarization in the monochromatic radiation field,[19,20] when polarizations of resonance allowed transitions interfere, the interference of polarizations of forbidden and quasi-resonance allowed transitions occurs in the above case.

The interference of polarizations at allowed and forbidden transitions has not been considered earlier in the literature. The predicted radiation amplification should be taken into account in quantitative measurements of absorption coefficients and forbidden transition oscillator strengths.

The influence of quasi-resonance polarizability on the field broadening reduces to an increase of saturation parameter κ (29), i.e., as is evident from the comparison of Eqs. (40) and (29), κ is multiplied by the coefficient $1 + w + w^2$, $\kappa \to (1 + w + w^2)\kappa$, which results in insignificant line widening at small extent of nonresonance allowed transition saturation at $w < 1$. The polarizability influences the line shift insignificantly.

It follows from Eq. (40) that an additional vertical (interference) line shift is Δw; its sign is determined by the sign of frequency detuning Δ. The vertical shift equals to the homogeneous half-width of line Γ when the intensity I_s is Δ/Γ-time lower than the intensity of nonresonance transition saturation, determined by the relation $w = 1$. For the above values, $\Delta = 1$ cm^{-1}, and $\Gamma = 100$ MHz $= 0.0033$ cm^{-1}, $I_s = c\hbar\Gamma\Delta/(2\pi d_2^2) = 67$ kW/cm^2.

Note, that the intensity I_s, determined from the condition $\Delta w/\Gamma = 1$, is equal to the intensity of saturation of stimulated Raman two-wavelength scattering[2] when changing $d_1 d_2$ to d_2^2 in the latter. If $I = 100$ MW/cm^2, then at $\Delta = 1$ cm^{-1} the vertical shift $\Delta w = 5$ cm^{-1} = 150 GHz, which is three order of magnitude larger than the homogeneous half-width of the line $\Gamma = 100$ MHz. At such intensity, the value of vertical shift is equal to Γ, i.e., vertical shift is quite noticeable, for quite significant frequency detuning: $\Delta \sim 10^3$–10^4 cm^{-1}.

Hence, the main influence of atom polarizability on absorption line profile at forbidden transitions in the quasi-resonance conditions $|\omega - \omega_{32}| \sim 10$–$1000\,\Gamma$ lies in formation of large vertical line shift.

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