

PACS numbers: 42.65.Dr; 42.50.Md
DOI: 10.1070/QE2009v039n02ABEH013926

Influence of phase memory effects in collisions on a resonance Raman spectrum

A.I. Parkhomenko, A.M. Shalagin

***Abstract*. The influence of phase memory effects on the spectrum of resonance Raman scattering by three-level atoms with the Λ configuration of levels experiencing collisions with buffer gas atoms in a strong monochromatic radiation field is studied theoretically. Systems with a small Doppler broadening compared to the collision frequency (large buffer gas pressures) are analysed in the general case of an arbitrary change (from complete change to complete preservation) in the phase memory at any of three transitions in the Λ system. It is shown that in the absence of the collision relaxation of the low-frequency coherence at the transitions between two lower levels of the Λ system, the radiation scattering spectrum has a spectrally narrow component at the Raman frequency, which, despite the homogeneous broadening of the absorption line, exhibits a strongly pronounced anisotropy. In the direction, close to the propagation direction of exciting radiation, this line maximally narrows down. It is significant that upon optical pumping to the level unaffected by a strong field the resonance Raman spectrum noticeably differs from the spectrum in the case of the probe field. A simple expression is proposed for calculating the degree of the phase memory preservation in collisions from the relative amplitude of the Raman resonance.**

Keywords: *resonance Raman scattering, collisions, coherence, populations of levels, spectrum, resonances, fluorescence.*

1. Introduction

Resonance scattering (resonance fluorescence, resonance Raman scattering) of strong monochromatic radiation by atoms and molecules has been actively studied for many years. Special attention in these studies is paid to a three-level Λ-system consisting of two close lower levels and the third remote level which is optically connected with two lower levels (see, for example, [1 – 5] and references therein). The effect of coherent trapping of populations is well pronounced in a three-level Λ system with long-lived lower levels and in fluorescence spectra it is observed in the form of a characteristic narrow dip ('dark' resonance) [4, 5].

A.I. Parkhomenko, A.M. Shalagin Institute of Automation and Electrometry, Siberian Branch, Russian Academy of Sciences, prosp. Akad. Koptyuga 1, 630090 Novosibirsk, Russia;
e-mail: par@iae.nsk.su, shalagin@iae.nsk.su

Received 10 June 2008
Kvantovaya Elektronika **39** (2) 163 – 170 (2009)
Translated by I.A. Ulitkin

The fluorescence spectrum of the upper level in the Λ system is commonly studied by using two light fields. The frequency of one (strong) field is fixed and the frequency of the other (probe) field acting on the adjacent transition is scanned [4, 5]. In theoretical papers the fluorescence spectrum in such systems was studied under some or other assumptions and approximations: calculations have been performed either by neglecting the motion and collisions of atoms or by using simplest relaxation models. At the same time, a complete and correct account for relaxation processes in calculating spontaneous emission spectra is very important. In this paper, we studied theoretically the spontaneous emission spectrum of three-level atoms with the Λ configuration experiencing collisions with buffer gas atoms in the field of a strong electromagnetic wave. It was assumed that only one external field affects the atom (probe field is absent) and spontaneous emission is detected at the transition adjacent with the transition perturbed by the external field. Because an open Λ system is considered, particles are not completely transferred to the lower level not excited by the external field. The analysis was performed for the general case of an arbitrary change (from complete change to complete preservation) in the phase memory in collisions at any three transitions in the Λ system.

2. Basic equations

Consider a gas of three-level absorbing particles in a mixture with a buffer gas. We will neglect collisions between absorbing particles by assuming that the concentration of the buffer gas is greater than that of the absorbing gas. Let three-level atoms with the Λ configuration (Fig. 1) be excited by a monochromatic field

$$\mathcal{E} = \mathrm{Re}\boldsymbol{E}\exp(\mathrm{i}\boldsymbol{k}\boldsymbol{r} - \mathrm{i}\omega t)$$

with the frequency ω close to the frequency ω_{mn} of the $m - n$ transition between m and n levels (here, $\boldsymbol{E}$ and $\boldsymbol{k}$ are the electric field strength and the radiation wave vector). The spontaneous decay of the m level over the $m \to n$ and $m \to l$ channels is characterised by the constants A_{mn} and A_{ml}. The escape of atoms from the region of interaction with the light beam (due to their motion and chemical reactions) and possible transitions of atoms from the m, n and l levels due to the spontaneous decay to other levels will be characterised by the relaxation constants γ_m, γ_n and γ_l. The transitions of atoms to the n and l levels (i.e. to the interaction region with the light beam because of their motion) are described by the pump rates $q_n(\boldsymbol{v})$ and $q_l(\boldsymbol{v})$, where $\boldsymbol{v}$ is the particle velocity.

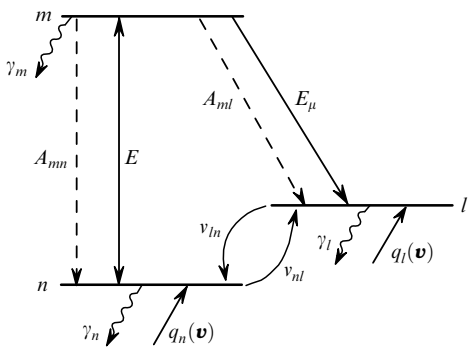

Figure 1. Energy level diagram of and transitions between levels.

We will describe spontaneous emission at the $m-l$ transition according to [1] by introducing the classic field E_μ. In this case, the calculation of the spontaneous emission spectrum is formally similar to the calculation of the spectrum in the case of the classical probe field E_μ, if in the latter calculation only emission is selected and E_μ is treated as the spectral density of zero oscillations of vacuum. Thus, to describe spontaneous emission at the $m-l$ transition, we use the kinetic equations for the elements of the density matrix [1]:

$$\begin{aligned}
&\left(\frac{\mathrm{d}}{\mathrm{d}t}+\Gamma_m\right)\rho_m(\boldsymbol{v}) = S(\rho_m(\boldsymbol{v}))+2\mathrm{Re}[\mathrm{i}G\rho_{nm}(\boldsymbol{v})],\\
&\left(\frac{\mathrm{d}}{\mathrm{d}t}+\gamma_n\right)\rho_n(\boldsymbol{v}) = S(\rho_n(\boldsymbol{v}))+A_{mn}\rho_m(\boldsymbol{v})+q_n(\boldsymbol{v})\\
&\quad -2\mathrm{Re}[\mathrm{i}G\rho_{nm}(\boldsymbol{v})],\\
&\left(\frac{\mathrm{d}}{\mathrm{d}t}+\gamma_l\right)\rho_l(\boldsymbol{v}) = S(\rho_l(\boldsymbol{v}))+A_{ml}\rho_m(\boldsymbol{v})+q_l(\boldsymbol{v})\\
&\quad +2\mathrm{Re}[\mathrm{i}G_\mu^*\rho_{ml}(\boldsymbol{v})],\\
&\left[\frac{\mathrm{d}}{\mathrm{d}t}+\frac{\Gamma_m+\gamma_n}{2}+\mathrm{i}(\Omega_0-\boldsymbol{k}\boldsymbol{v})\right]\rho_{nm}(\boldsymbol{v})\\
&\quad = S(\rho_{nm}(\boldsymbol{v}))-\mathrm{i}G^*[\rho_n(\boldsymbol{v})-\rho_m(\boldsymbol{v})],\\
&\left[\frac{\mathrm{d}}{\mathrm{d}t}+\frac{\Gamma_m+\gamma_l}{2}-\mathrm{i}(\Omega_{0\mu}-\boldsymbol{k}_\mu\boldsymbol{v})\right]\rho_{ml}(\boldsymbol{v})\\
&\quad = S(\rho_{ml}(\boldsymbol{v}))+\mathrm{i}G\rho_{nl}(\boldsymbol{v})-\mathrm{i}G_\mu\rho_m(\boldsymbol{v}),\\
&\left[\frac{\mathrm{d}}{\mathrm{d}t}+\frac{\gamma_n+\gamma_l}{2}-\mathrm{i}(\varepsilon_0-\boldsymbol{q}\boldsymbol{v})\right]\rho_{nl}(\boldsymbol{v})\\
&\quad = S(\rho_{nl}(\boldsymbol{v}))+\mathrm{i}G^*\rho_{ml}(\boldsymbol{v})-\mathrm{i}G_\mu\rho_{nm}(\boldsymbol{v}),
\end{aligned} \tag{1}$$

where

$$\begin{aligned}
&\Gamma_m = A_{nm}+A_{ml}+\gamma_m;\ \ \Omega_0=\omega-\omega_{mn};\\
&\Omega_{0\mu}=\omega_\mu-\omega_{ml};\quad \varepsilon_0=\Omega_{0\mu}-\Omega_0;\quad \boldsymbol{q}=\boldsymbol{k}_\mu-\boldsymbol{k};\\
&G=\frac{d_{mn}E}{2\hbar};\quad G_\mu=\frac{d_{mn}E_\mu}{2\hbar};\quad \frac{\mathrm{d}}{\mathrm{d}t}\equiv\frac{\partial}{\partial t}+\boldsymbol{v}\nabla;
\end{aligned} \tag{2}$$

$\rho_i(\boldsymbol{v})$ is the velocity distribution of particles at the i level ($i=m, n, l$); $S(\rho_i(\boldsymbol{v}))$ and $S(\rho_{ij}(\boldsymbol{v}))$ are collision integrals; d_{mn} and d_{ml} are matrix elements of dipole moments at the $m-n$ and $m-l$ transitions; ω_{ml} is the $m-l$ transition frequency; ω_μ is the frequency at which the spectral density of spontaneous emission is calculated; and $\boldsymbol{k}_\mu$ is the wave vector of spontaneous emission.

We assume below that quantities $q_i(\boldsymbol{v})$ in (1) are time independent and their dependence on $\boldsymbol{v}$ is Maxwellian:

$$\begin{aligned}
&q_i(\boldsymbol{v})=Q_iW(\boldsymbol{v}),\quad W(\boldsymbol{v})=\frac{\exp\left[-(\boldsymbol{v}/\upsilon_T)^2\right]}{\left(\sqrt{\pi}\upsilon_T\right)^3};\\
&\upsilon_T=\left(\frac{2k_\mathrm{B}T}{M}\right)^{1/2},\quad i=n, l,
\end{aligned} \tag{3}$$

where Q_i is the total (integrated over $\boldsymbol{v}$) pump rate to the i level; $W(\boldsymbol{v})$ is the Maxwell velocity distribution; k_B is the Boltzmann constant; M is the absorbing particle mass; T is the medium temperature; and υ_T is the most probable velocity of absorbing particles.

For collision integrals (1), we will use the model of strong collisions [1], taking into account the collision transitions between n and l levels:

$$\begin{aligned}
&S(\rho_m(\boldsymbol{v}))=-\nu_m\rho_m(\boldsymbol{v})+\nu_m\rho_mW(\boldsymbol{v}),\\
&S(\rho_n(\boldsymbol{v}))=-(\nu_n+\nu_{nl})\rho_n(\boldsymbol{v})+(\nu_n\rho_n+\nu_{ln}\rho_l)W(\boldsymbol{v}),\\
&S(\rho_l(\boldsymbol{v}))=-(\nu_l+\nu_{ln})\rho_l(\boldsymbol{v})+(\nu_l\rho_l+\nu_{nl}\rho_n)W(\boldsymbol{v}),\\
&S(\rho_{nl}(\boldsymbol{v}))=-\nu\rho_{nl}(\boldsymbol{v})+\tilde{\nu}\rho_{nl}W(\boldsymbol{v}),\\
&S(\rho_{nm}(\boldsymbol{v}))=-\nu_1\rho_{nm}(\boldsymbol{v})+\tilde{\nu}_1\rho_{nm}W(\boldsymbol{v}),\\
&S(\rho_{ml}(\boldsymbol{v}))=-\nu_2\rho_{ml}(\boldsymbol{v})+\tilde{\nu}_2\rho_{ml}W(\boldsymbol{v}),\\
&\rho_i\equiv\int\rho_i(\boldsymbol{v})\mathrm{d}\boldsymbol{v},\quad i=m, n, l, nl, nm, ml,
\end{aligned} \tag{4}$$

where ν_m, ν_n and ν_l are the frequencies of elastic collisions of absorbing particles in the states m, n, l with buffer particles; ν_{nl} and ν_{ln} are frequencies of collision transitions $n\to l$ and $l\to n$ in the Maxwell velocity distribution of particles; ν, ν_1, ν_2 and $\tilde{\nu}$, $\tilde{\nu}_1$, $\tilde{\nu}_2$ are 'nondiagonal' frequencies of the escape and arrival of particles, respectively, these frequencies being complex quantities in the general case. The condition

$$\tilde{\nu}=\tilde{\nu}_1=\tilde{\nu}_2=0 \tag{5}$$

corresponds to the case, when collisions cause complete relaxation of coherences $\rho_{nl}(\boldsymbol{v})$, $\rho_{nm}(\boldsymbol{v})$, $\rho_{ml}(\boldsymbol{v})$ (the absence of phase memory in collisions at all transitions). In the absence of the collision relaxation of coherence $\rho_{nl}(\boldsymbol{v})$ (at the $l-n$ transition the phase memory in collisions is preserved), collision transitions between n and l levels are absent ($\nu_{nl}=\nu_{ln}=0$) and the 'escape' (ν) and 'arrival' ($\tilde{\nu}$) frequencies are real and equal to each other [1]:

$$\tilde{\nu}=\nu=\nu_n=\nu_l\equiv\nu_{\mathrm{tr}}, \tag{6}$$

where v_{tr} means the average transport frequency of elastic collisions between active particles and buffer particles [6]. The quantity v_{tr} is related to the diffusion coefficient D of particles interacting with radiation by the expression $D = \upsilon_T^2/(2v_{tr})$ [7]. In the absence of the collision relaxation of coherences $\rho_{nm}(\boldsymbol{\upsilon})$ or $\rho_{ml}(\boldsymbol{\upsilon})$, relations $\tilde{v}_1 = v_1 = v_m = v_n$ and $\tilde{v}_2 = v_2 = v_m = v_l$ are fulfilled, respectively.

We will seek for the solution of Eqn (1) in the form

$$\rho_i(\boldsymbol{\upsilon}) = R_i(\boldsymbol{\upsilon}) + r_i(\boldsymbol{\upsilon}), \quad i = m, n, l, nl, nm, ml. \tag{7}$$

The matrix elements $R_i(\boldsymbol{\upsilon})$ correspond to the solution of the problem on the interaction with only one strong field E. Small additions $r_i(\boldsymbol{\upsilon})$ are caused by the presence of a weak field E_μ.

Under stationary and spatially uniform conditions, system of equations (1) after substitution of (7) into it in the first approximation with respect to G_μ is split to two subsystems:

$$\begin{aligned}
&(\Gamma_m + v_m)R_m(\boldsymbol{\upsilon}) = v_m R_m W(\boldsymbol{\upsilon}) + 2\mathrm{Re}[\mathrm{i}GR_{nm}(\boldsymbol{\upsilon})],\\
&(\gamma_n + v_n + v_{nl})R_n(\boldsymbol{\upsilon}) = A_{mn}R_m(\boldsymbol{\upsilon}) + Q_n W(\boldsymbol{\upsilon})\\
&\quad + (v_n R_n + v_{in}R_l)W(\boldsymbol{\upsilon}) - 2\mathrm{Re}[\mathrm{i}GR_{nm}(\boldsymbol{\upsilon})],\\
&(\gamma_l + v_l + v_{ln})R_l(\boldsymbol{\upsilon}) = A_{ml}R_m(\boldsymbol{\upsilon}) + Q_l W(\boldsymbol{\upsilon})\\
&\quad + (v_l R_l + v_{nl}R_n)W(\boldsymbol{\upsilon}),\\
&\left[\frac{\Gamma_m + \gamma_n}{2} + v_1 + \mathrm{i}(\Omega_0 - \boldsymbol{k}\boldsymbol{\upsilon})\right]R_{nm}(\boldsymbol{\upsilon})\\
&\quad = \tilde{v}_1 R_{nm}W(\boldsymbol{\upsilon}) - \mathrm{i}G^*[R_n(\boldsymbol{\upsilon}) - R_m(\boldsymbol{\upsilon})]
\end{aligned} \tag{8}$$

and

$$\begin{aligned}
&\left[\frac{\Gamma_m + \gamma_l}{2} + v_2 - \mathrm{i}(\Omega_{0\mu} - \boldsymbol{k}_\mu\boldsymbol{\upsilon})\right]r_{ml}(\boldsymbol{\upsilon})\\
&\quad = \tilde{v}_2 r_{ml}W(\boldsymbol{\upsilon}) + \mathrm{i}Gr_{nl}(\boldsymbol{\upsilon}) - \mathrm{i}G_\mu R_m(\boldsymbol{\upsilon}),\\
&\left[\frac{\gamma_n + \gamma_l}{2} + v - \mathrm{i}(\varepsilon_0 - \boldsymbol{q}\boldsymbol{\upsilon})\right]r_{nl}(\boldsymbol{\upsilon})\\
&\quad = \tilde{v}r_{nl}W(\boldsymbol{\upsilon}) + \mathrm{i}G^* r_{ml}(\boldsymbol{\upsilon}) - \mathrm{i}G_\mu R_{nm}(\boldsymbol{\upsilon}).
\end{aligned} \tag{9}$$

The quantities R_m, R_n, R_l, R_{nm}, r_{ml}, r_{nl} in (8) and (9) are the integrals with respect to velocities from quantities $R_m(\boldsymbol{\upsilon})$, $R_n(\boldsymbol{\upsilon})$, $R_l(\boldsymbol{\upsilon})$, $R_{nm}(\boldsymbol{\upsilon})$, $r_{ml}(\boldsymbol{\upsilon})$, $r_{nl}(\boldsymbol{\upsilon})$, respectively.

According to general rules [1], the probability of spontaneous emission w at the frequency ω_μ per one absorbing atom is determined by the expression

$$w = \frac{2}{N}\,\mathrm{Re}\left(\mathrm{i}G_\mu^* r_{ml}\right), \tag{10}$$

where $N \equiv R_n + R_l + R_m$ is the concentration of absorbing particles. Thus, according to the posed problem we should find the quantity r_{ml} from the system of equations (8), (9).

3. Weakly rate-selective interaction of atoms with radiation

The solution of system of equations (8), (9) in the general case of arbitrary relation between the homogeneous and Doppler widths of the absorption line leads to a cumbersome expression for the probability of spontaneous emission, which can be analysed only with the help of numerical methods. To simplify the problem, we will consider a weakly selective interaction of atoms with radiation with respect to rates, when the distribution of atoms in states m, n, l hardly differs from the Maxwellian distribution. This case corresponds to small Doppler widths $k\upsilon_T$, $k_\mu\upsilon_T$ compared to the transport collision frequency:

$$k\upsilon_T, k_\mu\upsilon_T \ll v_{tr}. \tag{11}$$

Condition (11) allows one to apply a very simple and efficient method for solving Eqns (8), (9) with respect to integral quantities R_m, R_n, R_l, R_{nm}, r_{ml}, r_{nl} relative to velocities, the so-called method of preliminary averaging with respect to velocities. This computation method based on averaging with respect to velocities in the kinetic equations for the matrix density is described in detail in paper [8]. By applying this procedure to Eqns (8) and (9) similarly to that in paper [8], we obtain

$$\begin{aligned}
&\Gamma_m R_m = 2\mathrm{Re}(\mathrm{i}GR_{nm}),\\
&(\gamma_n + v_{nl})R_n = A_{mn}R_m + Q_n + v_{ln}R_l - 2\mathrm{Re}(\mathrm{i}GR_{nm}),\\
&(\gamma_l + v_{ln})R_l = A_{ml}R_m + Q_l + v_{nl}R_n,\\
&(\Gamma + \mathrm{i}\Omega_1)R_{nm} = -\mathrm{i}G^*(R_n - R_m),\\
&(\Gamma_\mu - \mathrm{i}\Omega_\mu)r_{ml} = \mathrm{i}Gr_{nl} - \mathrm{i}G_\mu R_m,\\
&(\Gamma_1 - \mathrm{i}\varepsilon)r_{nl} = \mathrm{i}G^* r_{ml} - \mathrm{i}G_\mu R_{nm}.
\end{aligned} \tag{12}$$

Here, we introduced the following notations:

$$\begin{aligned}
\Gamma &= \frac{\Gamma_m + \gamma_n}{2} + \mathrm{Re}(v_1 - \tilde{v}_1)\\
&\quad + \mathrm{Re}\left[\frac{(k\upsilon_T)^2}{2}\left(\frac{\Gamma_m + \gamma_n}{2} + v_1 + \mathrm{i}\Omega_0\right)^{-1}\right],\\
\Gamma_\mu &= \frac{\Gamma_m + \gamma_l}{2} + \mathrm{Re}(v_2 - \tilde{v}_2)\\
&\quad + \mathrm{Re}\left[\frac{(k_\mu\upsilon_T)^2}{2}\left(\frac{\Gamma_m + \gamma_l}{2} + v_2 - \mathrm{i}\Omega_{0\mu}\right)^{-1}\right],\\
\Gamma_1 &= \frac{\gamma_n + \gamma_l}{2} + \mathrm{Re}(v - \tilde{v})\\
&\quad + \mathrm{Re}\left[\frac{(q\upsilon_T)^2}{2}\left(\frac{\gamma_n + \gamma_l}{2} + v - \mathrm{i}\varepsilon_0\right)^{-1}\right],
\end{aligned} \tag{13}$$

$$\Omega_1 = \Omega_0 + \mathrm{Im}(v_1 - \tilde{v}_1), \quad \Omega_\mu = \Omega_{0\mu} - \mathrm{Im}(v_2 - \tilde{v}_2),$$

$$\varepsilon = \Omega_\mu - \Omega, \quad \Omega = \Omega_0 + \mathrm{Im}(v - \tilde{v} - v_2 + \tilde{v}_2).$$

The quantities Γ, Γ_μ and Γ_1 in (13) depend on the degree of preservation of the phase memory in collisions at transitions $m-n$, $m-l$ and $l-n$, respectively. In the absence of the phase memory at all transitions [see expression (5)] due to condition (11) the relation $\Gamma, \Gamma_\mu, \Gamma_1 \gg kv_T$ is fulfilled. In the case of complete preservation of the phase memory at the $l-n$ transition [see expression (6)] and in the absence of the phase memory at transitions $m-n$ and $m-l$, relations $\Gamma_1 \ll kv_T$ and $\Gamma, \Gamma_\mu \gg kv_T$ are fulfilled. Quantities Γ, Γ_μ, Γ_1 also depend on frequency detunings of exciting and scattering radiation. However, in describing the spontaneous emission spectrum in cases under study, we can neglect this dependence. Indeed, in the absence of the phase memory at all transitions, the second terms in right-hand sides of expressions for Γ, Γ_μ and Γ_1 become main and, hence, in these expressions the last terms containing the dependence on Ω_0, $\Omega_{0\mu}$ and ε_0 can be neglected. When the phase memory is preserved at all transitions, last terms in expressions for Γ, Γ_μ, Γ_1 will be the main ones. However, their dependence on Ω_0, $\Omega_{0\mu}$ and ε_0 becomes significant only in the long wing of the spectral line, which we do not consider in this paper.

The standard solution of Eqns (13) leads to the expression for the spontaneous emission probability w:

$$w = \frac{2|G_\mu|^2}{N}\,\mathrm{Re}\,\frac{(\Gamma_1 - \mathrm{i}\varepsilon)R_m + \mathrm{i}GR_{nm}}{(\Gamma_1 - \mathrm{i}\varepsilon)(\Gamma_\mu - \mathrm{i}\Omega_\mu) + |G|^2}, \tag{14}$$

where

$$R_m = \frac{\varkappa N_n}{1 + \varkappa(A_{ml} + \tilde{\gamma}_m + \tilde{\gamma}_n)/\tilde{\gamma}_n}; \quad \mathrm{i}GR_{nm} = \frac{\Gamma_m(\Gamma - \mathrm{i}\Omega_1)}{2\Gamma}\,R_m;$$

$$\varkappa = \frac{2|G|^2}{\Gamma_m}\,\mathrm{Re}\,\frac{1}{\Gamma - \mathrm{i}\Omega_1}; \quad N_n = \frac{Q_n + (Q_n + Q_l)v_{ln}/\gamma_l}{\tilde{\gamma}_n}; \tag{15}$$

$$\tilde{\gamma}_m = \gamma_m + v_{ln}\gamma_m/\gamma_l; \quad \tilde{\gamma}_n = \gamma_n + v_{nl} + v_{ln}\gamma_n/\gamma_l.$$

The physical sense of quantities $\varkappa$ and N_n in (15) becomes clear from relations following from (12):

$$R_m = \frac{\varkappa}{1+\varkappa}\,R_n, \quad R_n = N_n\left(1 + \frac{A_{ml} + \tilde{\gamma}_m}{\tilde{\gamma}_n}\,\frac{\varkappa}{1+\varkappa}\right)^{-1}. \tag{16}$$

The quantity $\varkappa$, as one can see from the first relation in (16) characterises the degree of equalising populations of levels m and n, and therefore it can be interpreted as a saturation parameter for the $m-n$ transition. The quantity N_n in accordance with the second relation in (16) is the population of the n level in the absence of radiation (at $\varkappa = 0$). In a particular case

$$\gamma_m = \gamma_l = \gamma_n \equiv \gamma \tag{17}$$

Expression (15) for the quantity N_n takes the form

$$N_n = N\left(\frac{Q_n\gamma}{Q_n + Q_l} + v_{ln}\right)(\gamma + v_{nl} + v_{ln})^{-1}. \tag{18}$$

When the pump rates to the n and l levels ($Q_n = Q_l$) are equal and the collision exchange between these levels is weak (v_{nl}, $v_{ln} \ll \gamma$) half the absorbing particles is at the n level in the absence of radiation, $N_n = N/2$ as should be expected.

Note in conclusion to this section that expression (14) for the spontaneous emission probability is a particular case of the expression for the absorption probability P_μ of a probe field at the adjacent transition $m-l$. The expression for P_μ follows from Eqns (1) if they contain terms corresponding to the probe field absorption [one should add the terms $-2\mathrm{Re}[\mathrm{i}G_\mu^*\rho_{ml}(\boldsymbol{v})]$, $-\mathrm{i}G_\mu^*\rho_{nl}(\boldsymbol{v})$ and $\mathrm{i}G_\mu\rho_l(\boldsymbol{v})$ to the right-hand sides of the first, fourth and fifth expressions in (1)]. Under stationary and spatially uniform conditions for the absorption probabilities of the probe field we obtain the expression:

$$P_\mu = \frac{2|G_\mu|^2}{N}\,\mathrm{Re}\,\frac{(\Gamma_1 - \mathrm{i}\varepsilon)(R_l - R_m) - \mathrm{i}GR_{nm}}{(\Gamma_1 - \mathrm{i}\varepsilon)(\Gamma_\mu - \mathrm{i}\Omega_\mu) + |G|^2}, \tag{19}$$

where

$$R_l = \frac{Q_n + Q_l}{\gamma_l} - \left(\frac{\gamma_m}{\gamma_l} + \frac{1+\varkappa}{\varkappa}\,\frac{\gamma_n}{\gamma_l}\right)R_m \tag{20}$$

is the population of the l level. Thus, spontaneous emission probability w is described by expression (19) with the quantity R_l being excluded. Expression (19) for the absorption probability of the probe field is a natural generalisation of the corresponding expression for particles at rest {see, for example, expression (8.77) in [1]} and coincides with it at $v_T = 0$. The motion of atoms is manifested only in the change of relaxation constants Γ_1, Γ_μ, Γ due to additions reflecting the diffusion law of the particle transfer.

Let us emphasise the following important circumstance. In this paper we will consider the spontaneous emission spectrum under conditions of significant (but not complete) optical transfer of particles to the lower level not excited by the external field (at $R_l \gg R_n$, R_m). Under these conditions, the spontaneous emission spectrum strongly differs from the probe field spectrum. Indeed, under conditions of strong optical pumping of particles to the l level in expression (19), we should put $R_l \approx N$ and quantities R_m and R_{nm} can be neglected (the probe field spectrum under these conditions is considered in [9]). Under the same conditions the term in expression (14), which is proportional to the external-radiation-induced coherence R_{nm} of states m and n, is significant and cannot be neglected. This causes considerable difference of the spontaneous emission spectrum from the probe field spectrum.

4. Analysis of the resonance light scattering spectrum

Let us analyse expression (14) for the spontaneous emission probability. The denominator in (14) is quadratic with respect to the frequency detuning Ω_μ, i.e. the spontaneous emission spectrum has two resonances (two spectral components). This circumstance reflects the effect of field splitting of the m level into two quasi-energy levels. The position of the resonances can be easily determined by expanding the denominator in (14) to the product of two linear factors. In the case of noticeable splitting of levels ($\Omega_\mathrm{R} \gg |\Gamma_\mu - \Gamma_1|$) the result is known (see, for example, [1–3]): the maxima of the spectral components are located in the vicinity of $\Omega_\mu = \Omega_\mu^\pm$, where

$$\Omega_\mu^\pm = \frac{1}{2}(\Omega \pm \Omega_\mathrm{R}), \quad \Omega_\mathrm{R} \equiv \left(4|G|^2 + \Omega^2\right)^{1/2}. \tag{21}$$

According to (21) the distance between the maxima of the spectral components is equal to the generalised Rabi frequency Ω_R.

The nondiagonal element R_{nm} in (14) characterises the coherence of the states m and n, which is induced by a strong external field. In expression (14) the term proportional to R_{nm} describes the change in the line shape of scattered radiation and rather than its integrated intensity because

$$\int_{-\infty}^{+\infty} w\,\mathrm{d}\Omega_\mu = 2\pi|G_\mu|^2\,\frac{R_m}{N}. \tag{22}$$

Thus, the integrated intensity of scattered radiation is affected by a strong field only via the population of the m level.

It is convenient to represent expression (14) for the spontaneous emission probability in the form

$$w = 2|G_\mu|^2\,\frac{R_m}{N}\,B_w\,, \tag{23}$$

$$B_w = \mathrm{Re}\left\{\left[\Gamma_1 - \mathrm{i}\varepsilon + \frac{\Gamma_m(\Gamma - \mathrm{i}\Omega_1)}{2\Gamma}\right] \times\left[(\Gamma_1 - \mathrm{i}\varepsilon)(\Gamma_\mu - \mathrm{i}\Omega_\mu) + |G|^2\right]^{-1}\right\}.$$

The factor B_w in (23) describes the shape of the spontaneous emission spectrum. Figures 2–4 show the spectra of resonance radiation scattering calculated by (23) at different intensities (it is proportional to $|G|^2$), the frequency detunings Ω of pump radiation and angles θ between the wave vectors $\boldsymbol{k}_\mu$ and $\boldsymbol{k}$. The spectra were calculated for the most interesting cases of complete preservation of the phase memory in collisions at the $l-n$ transition ($\tilde{\nu} = \nu$, $\nu_{ln} = \nu_{nl} = 0$, in this case $\Gamma_1 \ll kv_T$) and in the absence of the phase memory at the $m-n$ and $m-l$ transitions ($\tilde{\nu}_1 = \tilde{\nu}_2 = 0$, in this case Γ, $\Gamma_\mu \gg kv_T$). The relaxation constants γ_m, γ_n и γ_l were assumed equal [see expression (17)], which always holds true when atoms escape from the region of their interaction with a light beam due to their motion (levels m, n and l do not decay into other levels). To normalise the spectra in all figures, we used the quantity w_0, which implies the spontaneous emission probability at the line centre at the $m-l$ transition in the absence of pumping and under the conditions that the population R_m of the m level from which emission occurs remains the same as that in the presence of pumping:

$$w_0 = \frac{2|G_\mu|^2}{\Gamma_\mu}\,\frac{R_m}{N}. \tag{24}$$

One can see from Figs 2–4 that the spontaneous emission spectrum exhibits two spectral components: a broad one in the vicinity of the frequency of the $m-l$ transition (near $\Omega_\mu \approx 0$) and a narrow one in the vicinity of the Raman frequency (near $\Omega_\mu \approx \Omega$). The broad component is isotropic with respect to the mutual orientation of wave vectors of strong and scattered radiation. The narrow component, despite the homogeneous broadening [Doppler broadening is small compared to the collision frequency, see expression (11)], exhibits a strongly pronounced anisotropy. This line in the scattering spectra narrows down maximally in the direction close to the propagation direction of exciting radiation ($\theta = 0$). The spectrum of scattered radiation depends on the exciting radiation intensity (cf. Figs 2a, 3a and 4a).

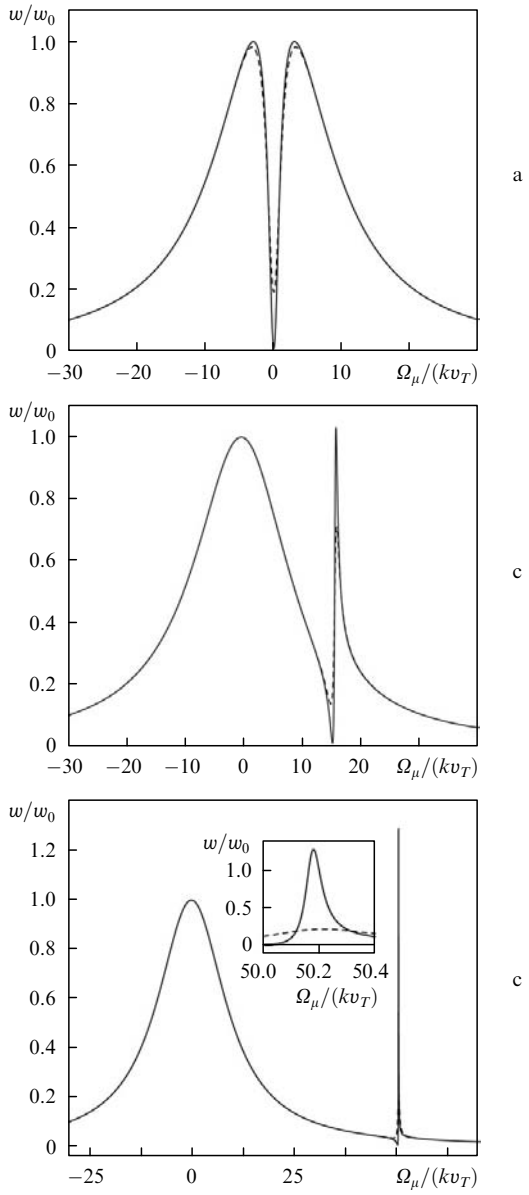

Figure 2. Spectra of resonance radiation scattering by three-level Λ atoms in the case of complete preservation of the phase memory in collisions at the $l-n$ transition ($\tilde{\nu} = \nu$, $\nu_{ln} = \nu_{nl} = 0$, $\Gamma_1 \ll kv_T$) and in the absence of the phase memory at the $m-n$ and $m-l$ transitions ($\tilde{\nu}_1 = \tilde{\nu}_2 = 0$, Γ, $\Gamma_\mu \gg kv_T$) for $\nu_{tr}/(kv_T) = 10$, $A_{mn}/(kv_T) = 10^{-2}$, $A_{ml} = A_{mn}$, $\gamma/A_{mn} = 10^{-4}$, $Q_n = Q_l$, $(k-k_\mu)/k = 10^{-4}$, $|G|/(kv_T) = 3$ ($2|G|^2 = 90\Gamma_\mu\Gamma_m$), $\theta = 0$ (solid curves) and π (dashed curves) $\Omega = 0$ (a), $\Omega/(kv_T) = 15$ (b) and 50 (c). The inset shows at an enlarged scale the resonance in the vicinity of $\Omega_\mu \approx \Omega$.

Note that only when the exciting radiation intensity is not high and the spectral components are not overlapped (at large frequency detunings of exciting radiation, $|\Omega| \gg \Gamma$), we can speak of some lines in the fluorescence and Raman spectra (correspondingly near $\Omega_\mu \approx 0$ and $\Omega_\mu \approx \Omega$). At a high radiation intensity, especially under the resonance conditions (for $|\Omega| < \Gamma$), two-photon and step processes are not independent, and we deal with resonance Raman scattering. This is especially obvious in the case of an exact resonance ($\Omega = 0$) and a high intensity ($2|G| \gg \Gamma_\mu, \Gamma_1$) of

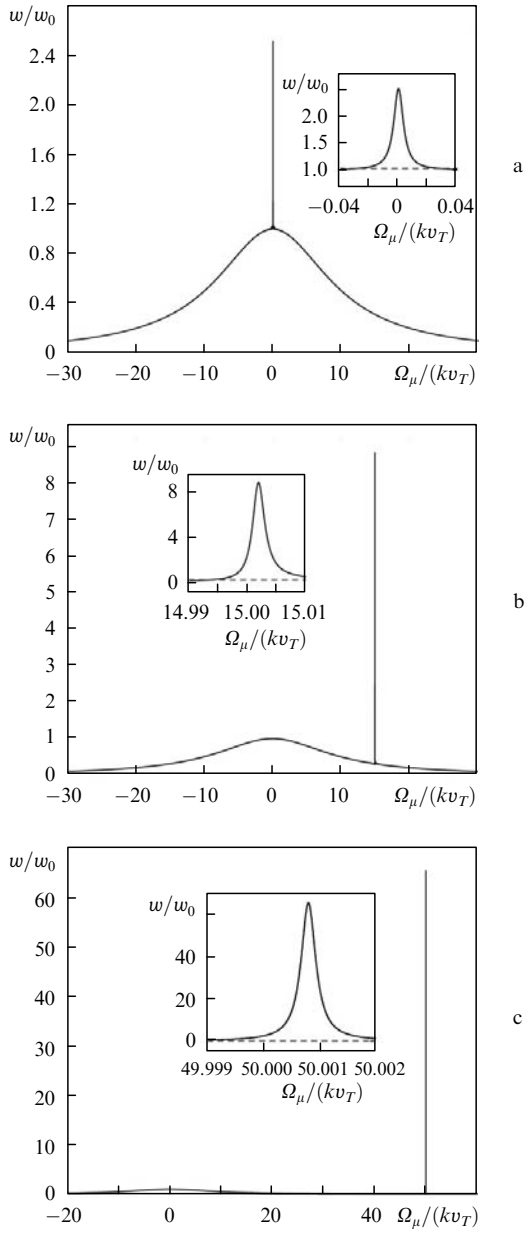

Figure 3. Spectra of resonance radiation scattering by three-level Λ atoms for $|G|/(kv_T) = 0.2$ ($2|G|^2 = 0.4\Gamma_\mu\Gamma_m$), $\Omega = 0$ (a), $\Omega/(kv_T) = 15$ (b) and 50 (c); other parameters and notations are the same as in Fig. 2.

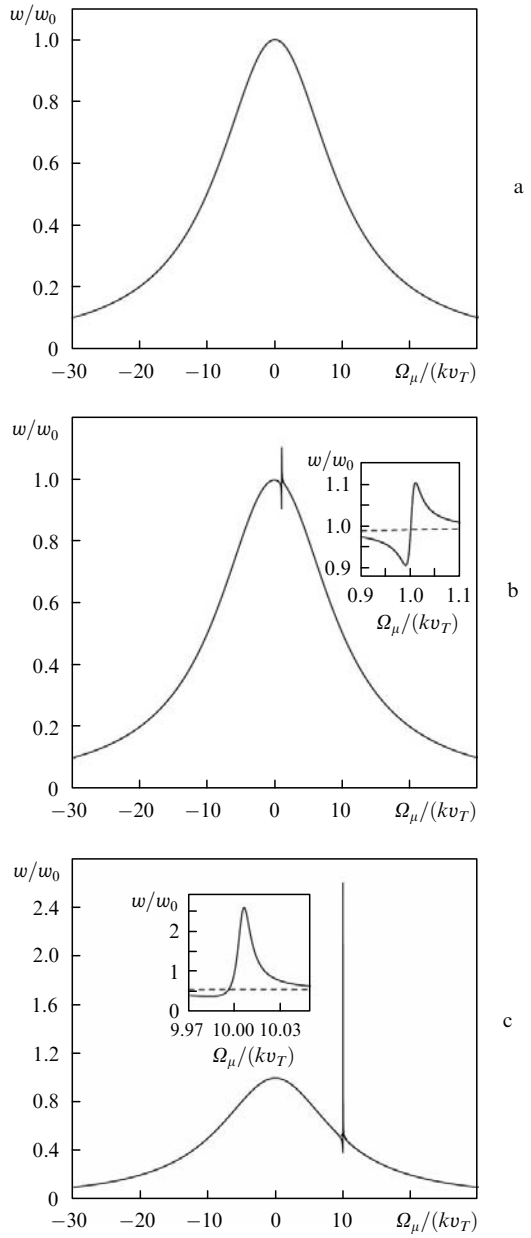

Figure 4. Spectra of resonance radiation scattering by three-level Λ atoms for $|G|/(kv_T) = 0.317$ ($2|G|^2 = \Gamma_\mu\Gamma_m$), $\Omega = 0$ (a), $\Omega/(kv_T) = 1$ (b) and 50 (c); other parameters and notations are the same as in Fig.2.

exciting field, when the scattered radiation spectrum is given by the expression [in this case, it follows from expression (23)]

$$B_w = \frac{1}{2}\,\mathrm{Re}\left[\frac{1}{\Gamma_{\mu 1} - \mathrm{i}(\Omega_\mu + |G|)} + \frac{1}{\Gamma_{\mu 1} - \mathrm{i}(\Omega_\mu - |G|)}\right], \qquad \Gamma_{\mu 1} = \frac{\Gamma_\mu + \Gamma_1}{2}. \tag{25}$$

One can see from (25) that in the case of the exact resonance and a high intensity of the exciting field the scattered radiation spectrum is symmetric with respect to $\Omega_\mu = 0$ and both spectral components are indiscernible in the amplitude and width (they are equal).

Consider in detail different regions of the scattered radiation spectrum. Let the frequency detuning and the intensity of exciting radiation be small

$$|\Omega| \ll \Gamma, \Gamma_\mu, \quad 2|G| \ll \Gamma_\mu, \tag{26}$$

and the relaxation constants of coherences at the $m - l$ and $l - n$ transitions differ significantly

$$\Gamma_1 \ll \Gamma_\mu \tag{27}$$

(collisions rather well preserve the phase memory at the $l - n$ transition). Under these conditions we obtain from (23)

$$B_w = \mathrm{Re}\left\{\frac{1}{\Gamma_\mu - \mathrm{i}\Omega_\mu} - \left(\frac{|G|^2}{\Gamma_\mu^2} - \frac{\Gamma_m}{2\Gamma_\mu}\right)[\Gamma_{\mathrm{eff}} - \mathrm{i}(\Omega_\mu - \Omega)]^{-1}\right\}, \qquad \Gamma_{\mathrm{eff}} = \Gamma_1 + \frac{|G|^2}{\Gamma_\mu}. \tag{28}$$

According to (28) the scattered radiation spectrum exhibits two spectral components of the Lorenz shape: at the

frequency of the $m-l$ transition ($\Omega_\mu = 0$, the first term in braces) and at the Raman frequency ($\Omega_\mu = \Omega$, the second term). The half-widths Γ_μ and Γ_{eff} of the corresponding components strongly differ ($\Gamma_\mu \gg \Gamma_{\text{eff}}$). The broad component (with the half-width Γ_μ) is isotropic with respect to the mutual orientation of the wave vectors $\boldsymbol{k}$ and $\boldsymbol{k}_\mu$ of exciting and scattered radiation, while the narrow components (with the half-width Γ_{eff}) is strongly anisotropic (Figs 2a and 3a). The anisotropy is most strongly pronounced at a low intensity of exciting radiation ($|G|^2 \ll \Gamma_\mu\Gamma_1$), when the half-width Γ_{eff} of the narrow component is equal to Γ_1:

$$\Gamma_{\text{eff}} \simeq \Gamma_1 = \frac{\gamma_n + \gamma_l}{2} + q^2 D. \tag{29}$$

The expression for Γ_1 in (29) is valid under condition (6) and when the inequality $\gamma_n + \gamma_l \ll 2v_{\text{tr}}$ is known to be fulfilled. If $\gamma_n + \gamma_l \ll 2q^2 D$, the half-width of the narrow anisotropic component in the scattered radiation spectrum proves to be proportional to the diffusion coefficient:

$$\Gamma_{\text{eff}} \simeq q^2 D. \tag{30}$$

This circumstance can be used to measure the diffusion coefficient of absorbing particles in the atmosphere of the buffer gas employing spectroscopic data.

The narrow anisotropic component is manifested in the scattered radiation spectrum as a dip or a peak against the background of a broad band depending on the exciting radiation intensity. There is a dip ('dark' resonance) against the background of the broad band, if $|G|^2 > \Gamma_m\Gamma_\mu/2$ (Fig. 2a) or a peak, if $|G|^2 < \Gamma_m\Gamma_\mu/2$ (Fig. 3a). At the given intensity of exciting radiation so that $|G|^2 = \Gamma_m\Gamma_\mu/2$, the narrow component in the scattered radiation spectrum is absent (Fig. 4a). Note that in the open Λ system the 'dark' resonance in the scattered radiation spectrum (Fig. 2a) appears under the action of only one external field.

Consider now the case of ultimately large frequency detuning of exciting radiation,

$$|\Omega| \gg \Gamma, \tag{31}$$

at its moderately high intensity, so that

$$|\Omega|\Gamma_1 \ll |G|^2 \ll |\Omega|^2. \tag{32}$$

In this case, expression (23) for the scattered radiation spectrum can be transformed to the form:

$$B_w = \left(1 - \frac{\Gamma_m}{2\Gamma}\right)\Gamma_\mu\left[\Gamma_\mu^2 + \left(\Omega_\mu + \frac{|G|^2}{\Omega}\right)^2\right]^{-1} + \left(\frac{|G|^2}{\Omega^2} + \frac{\Gamma_m}{2\Gamma}\right)(\Gamma_1 + \gamma_1)\left[(\Gamma_1 + \gamma_1)^2 + \left(\Omega_\mu - \Omega - \frac{|G|^2}{\Omega}\right)^2\right]^{-1}, \quad \gamma_1 = \frac{|G|^2\Gamma_\mu}{\Omega^2}. \tag{33}$$

The first term in (33) describes the shape of the fluorescence line, which is in the vicinity of $\Omega_\mu = \Omega_\mu^- \approx -|G|^2/\Omega$ and has the half-width Γ_μ. The second term describes the Raman line shape, which is in the vicinity of $\Omega_\mu = \Omega_\mu^+ \approx \Omega + |G|^2/\Omega$ and has the half-width $\Gamma_1 + \gamma_1$. The fluorescence line is isotropic with respect to the mutual orientation of the wave vectors $\boldsymbol{k}$ и $\boldsymbol{k}_\mu$ of exciting and scattered radiation, while the Raman line is anisotropic. The anisotropy is most strongly manifested when the phase memory at the $n-l$ transition is preserved (see insets in Figs 2c and 3c). In this case, the quantity Γ_1 is determined by expression (29) and, hence, the half-width $\Gamma_1 + \gamma_1$ of the Raman line depends on the diffusion coefficient D of particles interacting with radiation. If the condition

$$\gamma_n, \gamma_l, \gamma_1 \ll q^2 D \tag{34}$$

is fulfilled, the half-width of the Raman line is proportional to the diffusion coefficient:

$$\Gamma_1 + \gamma_1 \simeq q^2 D. \tag{35}$$

One can see from (33) that the ratio of the amplitude of Raman resonance to the amplitude of the fluorescence resonance is

$$A_{\text{R}} = \left(\gamma_1 + \frac{\Gamma_m}{2}\frac{\Gamma_\mu}{\Gamma}\right)\left[(\Gamma_1 + \gamma_1)\left(1 - \frac{\Gamma_m}{2\Gamma}\right)\right]^{-1}. \tag{36}$$

In the absence of the phase memory at all transitions (in this case, $\Gamma_1 \approx \Gamma_\mu \approx \Gamma \gg \gamma_1, \Gamma_m$), the relative amplitude of the Raman resonance is small ($A_{\text{R}} \ll 1$). The preservation of the phase memory at the $n-l$ transition (in this case, $\Gamma_\mu \approx \Gamma \gg \Gamma_1, \gamma_1, \Gamma_m$) leads to a drastic increase in the amplitude of the Raman resonance and it can be many times higher than the amplitude of the fluorescence resonance ($A_{\text{R}} \gg 1$). If condition (34) is fulfilled, the relative amplitude achieves its maximum value:

$$A_{\text{R}} = \frac{\Gamma_m}{2q^2 D}\frac{\Gamma_\mu}{\Gamma} \approx \frac{\Gamma_m}{2q^2 D}. \tag{37}$$

Thus, when the phase memory is preserved at the $n-l$ transition and condition (34) is fulfilled, the relative amplitude of the Raman resonance is inversely proportional to the diffusion coefficient D of particles interacting with radiation, i.e. it increases with increasing the buffer gas pressure.

When the phase memory is preserved partially (not completely) at the $n-l$ transition, the degree of its preservation is conveniently characterised by the parameter $\text{Re}\,\tilde{\nu}/\text{Re}\,\nu$ ($0 \leqslant \text{Re}\,\tilde{\nu}/\text{Re}\nu \leqslant 1$). This parameter can be found using the relative amplitude A_{R} (36) of the Raman resonance. Indeed, at a partial preservation of the phase memory at the $n-l$ transition, the second term in (13) for Γ_1 becomes the main one, so that

$$\Gamma_1 = \text{Re}(\nu - \tilde{\nu}). \tag{38}$$

Under the condition $\gamma_1 \ll \Gamma_1$ we obtain from expressions (36) and (38) a formula relating the relative amplitude A_{R} of the Raman resonance with the degree of phase memory preservation in collisions:

$$\frac{\text{Re}\,\tilde{\nu}}{\text{Re}\,\nu} = 1 - \frac{\Gamma_m}{2A_{\text{R}}\text{Re}\,\nu}\frac{\Gamma_\mu}{\Gamma} \approx 1 - \frac{\Gamma_m D}{v_T^2 A_{\text{R}}}\frac{\Gamma_\mu}{\Gamma}. \tag{39}$$

The second approximate equality in (39) (with the diffusion coefficient D in the right-hand side) is in fact precise at a high enough degree of the phase memory preservation at the $n-l$ transition (for $1 - \text{Re}\,\tilde{\nu}/\text{Re}\,\nu \ll 1$) because in this case we can assume [see expression (6)]

$$\text{Re}\,\nu = \nu_{\text{tr}} = v_T^2/(2D).$$

It follows from (33) that the ratio of the radiation energy in the Raman resonance to the energy on the fluorescence resonance (energies are integrals with respect to the frequency)

$$E_{\mathrm{R}} = \left(\frac{|G|^2}{\Omega^2} + \frac{\Gamma_m}{2\Gamma}\right)\left(1 - \frac{\Gamma_m}{2\Gamma}\right)^{-1}. \quad (40)$$

In this expression the factor $\Gamma_m/(2\Gamma)$ is caused by the exciting-radiation-induced coherence R_{nm} of the states m and n. It characterises the relaxation probability of the coherence R_{nm} in the radiation channel. If the collisions do not preserve the phase memory at the $m-n$ transition, this factor is small [$\Gamma_m/(2\Gamma) \ll 1$] and the energy of the Raman line is much lower that that of the fluorescence line $E_{\mathrm{R}} \ll 1$. If the phase memory at the $m-n$ transition is preserved, the factor $\Gamma_m/(2\Gamma)$ is close to unity and almost all the energy will be confined in the Raman line ($E_{\mathrm{R}} \gg 1$)).

5. Conclusions

We have studied theoretically the influence of phase memory effects in collisions on the spectrum of the resonance Raman scattering of radiation by three-level atoms with the Λ configuration experiencing collisions with the buffer gas atoms in a strong field of monochromatic radiation. The spectrum of resonance radiation scattering has been studied at the transition adjacent with the transition perturbed by the external field. The case of the homogeneous broadening of the absorption line has been analysed for large enough buffer gas pressures when the collision frequency is large compared to the Doppler width of the absorption line ($\nu_{\mathrm{tr}} \gg kv_T$).

The scattered radiation spectrum has two spectral components. When the intensity of exciting radiation is not high and these components are not overlapped (at a large frequency detuning of exciting radiation, $|\Omega| \gg \Gamma$), we deal with separate fluorescence and Raman lines near $\Omega_\mu \approx 0$ and $\Omega_\mu \approx \Omega$, respectively). Under resonance conditions (for $|\Omega| < \Gamma$) two-photon and step processes are not independent and we should speak of resonance Raman scattering.

It has been shown that the most interesting features appear in the scattered radiation spectrum in the absence of the collision relaxation of the low-frequency coherence at the transition between two lower levels of the Λ system. It has turned out that in this case the spectral component in the vicinity of the Raman frequency drastically narrows down and, despite the homogeneous broadening of the absorption line, has a strongly pronounced anisotropy with respect to the mutual orientation of the wave vectors of exciting and scattered radiation (maximum narrowing occurs in the direction close to the propagation direction of exciting radiation). In addition, the width and amplitude of the narrow anisotropic component depend on the diffusion coefficient $D = v_T^2/(2\nu_{\mathrm{tr}})$ of particles absorbing radiation. Thus, the diffusion coefficient of absorbing particles in the buffer gas atmosphere can be measured using spectroscopic data. A simple formula has been obtained, which can be used to find the parameter $\mathrm{Re}\,\tilde{\nu}/\mathrm{Re}\,\nu$ – the degree of phase memory preservation in collisions at the $n-l$ transition – by the relative amplitude of the Raman scattering resonance.

Under the conditions of significant optical transfer of particles to the lower level unaffected by the external field ($R_l \gg R_n,\ R_m$), the spectrum of the resonance Raman scattering differs considerably from the probe field spectrum (the probe field spectrum under these conditions is described in paper [9]). The spectra coincide only at a small frequency detuning ($|\Omega| \ll \Gamma,\ \Gamma_\mu$) and a high enough intensity ($|G|^2 \gg \Gamma_m\Gamma_\mu/2$) of exciting radiation, when a dip observed against the broad-band background (Fig. 2a). At a small intensity ($|G|^2 < \Gamma_m\Gamma_\mu/2$) and exact resonance for exciting radiation ($\Omega = 0$), the Raman spectrum exhibits a peak (Fig. 3a), while the probe field spectrum would exhibit a dip [9]. In the case of a large frequency detuning of exciting radiation ($|\Omega| \gg \Gamma$), the amplitude of the narrow Raman resonance is many times higher than the amplitude of the fluorescence resonance (Fig. 3c), whereas in the probe field spectrum the amplitude of the narrow resonance in the absorption line wing does not exceed the resonance amplitude near the centre of this line.

The peculiarities of the scattered radiation spectrum mentioned in this paper are most strongly pronounced in the case of a high enough degree of phase memory preservation in collisions at the $n-l$ transition (for $1 - \mathrm{Re}\,\tilde{\nu}/\mathrm{Re}\,\nu \ll 1$). For alkali metal atoms (they are well simulated by the Λ scheme of levels) in the atmosphere of inert buffer gases, the cross section of collision transitions between n, l components of the superfine structure of the ground state is 6–10 orders of magnitude smaller than the gas-kinetic cross sections [10]. Thus, these objects should exhibit a high degree of phase memory preservation in collisions so that $1 - \mathrm{Re}\,\tilde{\nu}/\mathrm{Re}\,\nu \lesssim 10^{-6}$. Therefore, alkali metal atoms in the atmosphere of inert gases are suitable enough to find and study all the peculiarities of the scattered radiation spectrum considered in this paper.

Acknowledgements. This work was supported by the Russian Foundation for Basic Research (Grant No. 07-02-00954), the program of the Department of Physical Sciences, Russian Academy of Sciences 'Optical spectroscopy and frequency standards' and by the Grant of the RF President for the State Support of Leading Scientific Schools (Grant No. NSh-1527.2008.2).

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