Double-resonance spectroscopy by a transversely nonuniform pump beam in a three-level molecular system

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We present a theoretical analysis of the influence of the collimation of a strong pump beam on a small-signal-gain line shape in a three-level Λ-type molecular system in the regime of high pump intensity when saturation and Stark (Autler-Townes) splitting of the levels of the pumping transition take place. The cases of homogeneous and Doppler-broadened pumping transitions have been considered. A significant offset, an asymmetric deformation, and a narrowing of the gain line, i.e., a spectrum focusing, have been obtained and explained as a result of the nonlinear (dispersion) and the active (gain) wave guiding of the small signal in the field of the strong pump beam.

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I. INTRODUCTION

Focusing or collimation of the pumping laser beam is usually used in schemes of nonlinear resonance spectroscopy when small-signal amplification is performed. The nonuniform intensity profile of the pump beam induces a nonuniform transverse distribution for the gain and for the nonlinear refractive index of the medium. Propagation of an electromagnetic wave in the case of a transverse quadratic gain or (and) dispersion variation has been analyzed in [1–4]. The bell-like square hyperbolic secant function for the transverse intensity distribution of the pump beam has been applied in [5] where the process of the simulated Raman scattering (SRS) has been investigated. As was shown later, this transverse intensity distribution corresponds to the spatial soliton in a nonlinear medium [6–8]. A number of authors have dealt with pump-beam focusing [9–11]. The SRS of a spatially nonuniform pump beam has been investigated by taking into account the depletion of the pump beam in [12] and by including Stokes–anti-Stokes interaction in [13].

The wave-guiding properties of the system induced by the transverse variation of the gain and the nonlinear refractive index due to the transverse variation of the intensity of the strong pump beam depend on the frequency of the small signal and spatial parameters, and the intensity and frequency of the pump beam. Therefore, one expects a change of the gain line shape of a small signal with respect to the case when a plane transversely homogeneous pump wave has been applied. An offset to the higher Stokes frequencies and a deformation of the Raman gain line were obtained earlier for homogeneously broadened quantum transitions in [4,5,14]. Neither the saturation of the material system or the Stark splitting of the levels of the transitions had been taken into account.

The present paper addresses an important practical problem of the small-signal-gain line-shape investigation in a three-level vibrational-rotational molecular system in the field of a strong collimated pump beam when the saturation and dynamic Stark (Autler-Townes) splitting of the levels of the pumping transition play an important role. Molecular three-level Λ-type systems with both homogeneously and Doppler-broadened pumping transitions are considered.

For infrared–far-infrared double-resonance spectroscopic [15] or Dicke superradiance [16] investigations, pumping by a laser working in the infrared is usually used. The pumping light induces a vibrational-rotational transition when the amplification takes place at the transition between rotational levels of the vibrationally excited manifold. Thus, the wavelengths of the pump and the amplified radiation may differ to a considerable extent. This causes different diffraction properties for the pump and the amplified signal beam. In this situation, it is possible to neglect the diffraction of the pump beam along the length of the gain medium when the diffraction of the amplified signal must be taken into account. The other possibility of neglecting the diffraction of the strong pump beam consists in the assumption that the pump beam represents a spatial soliton [6–8], which propagates without changing its intensity profile. We show that the small-signal-gain line shape obtained for the transversely nonuniform collimated pump beam in the Λ-type three-level molecular system, taking into account the transition saturation and the dynamic Stark (Autler-Townes) effect on the pump transition, can significantly differ from that for a plane uniform pump wave. An offset, deformation, and significant narrowing (referred to here as the effect of spectrum focusing of the signal) of the gain line shape, as well as the appearance of an absorption region in it, have been obtained and analyzed. These properties have been interpreted as a result of the influence of an active waveguide induced by the pump beam in the nonlinear gain medium with saturation and Stark splitting of the levels of the pump transition. The nonlinear wave guiding occurs only for some frequency regions of the signal and is absent for the other ones where only leaky modes can exist and where the absorption of the signal instead of the amplification takes place as a result of the diffraction losses. These frequency regions are determined by the transverse distribution of the pump intensity, pump and signal frequencies, and the gain coefficient of the medium. When an offset of the gain-line center to the higher Stokes frequencies has been obtained in the case of SRS in media with homogeneously broadened unsaturated transitions with no Stark splitting of the levels [4,5,14], the possibility of an offset in both directions, i.e., to the higher signal frequencies and to the lower ones as well, has been shown in the present paper. Note that some previous results of the investigation of the spectrum focusing effect are briefly presented in our paper [17].

The paper is organized as follows: In Sec. II we formulate the Bloch equations for the density-matrix elements of the Λ-type three-level molecular system and the wave equation for the amplitude of the small signal in the paraxial approximation. In Sec. III we solve these equations for the case of homogeneously broadened quantum transitions when collimation of the pump beam with Gaussian transverse intensity distribution has been applied. The case of Doppler-broadened pumping and homogeneously broadened signal transitions is discussed in Sec. IV. Finally, the results obtained in this paper are summarized in Sec. V, and possible applications of the spectrum focusing effect are considered.

II. THE MATHEMATICAL FORMALISM

We begin with the Bloch equations for the density-matrix elements ρ_{mn} of the three-level molecular system (Fig. 1) and with the wave equation in the paraxial approximation for the slowly varying complex amplitude A_S of the Stokes wave (the amplitude A_p of the strong pump wave is assumed as a given function of space and time coordinates) [18]:

$$\dot{\rho}_{mn}+\rho_{mn}/T_{mn}=(i/\hbar)[\rho,H]_{mn}\ ,$$
$$\dot{\rho}_{nn}+(\rho_{nn}-\rho^0_{nn})/T_{nn}=(i/h)[\rho,H]_{nn}$$
$$(m,n=1,2,3)\ , \qquad (1)$$
$$\left[\frac{\partial}{\partial z}+\frac{i}{2k_S}\Delta_\perp\right]A_S=-if_S(\varepsilon_1,\varepsilon_2,|A_p(x,y)|)A_S\ .$$

Here the Hamiltonian is taken to be $\hat{H}=\hat{H}_0-\hat{d}E(t,\mathbf{r})$, with $\hat{d}$ being the projection of the operator of the dipole moment on the direction of polarization of the total electric field $\mathbf{E}(t,\mathbf{r})$:

$$\mathbf{E}(t,z)=\tfrac{1}{2}\{\,\mathbf{A}_p\exp[i(\omega_p t-k_p z)]+\mathbf{A}_s\exp[i(\omega_s t-k_s z)]+\text{c.c.}\}\ .$$

Linear polarization in the same direction is assumed for the pump and Stokes waves for the sake of simplicity. ρ_{mn} and ρ_{nn} are off-diagonal and diagonal density-matrix elements. The latter ones represent populations of the corresponding levels. T_{mn} and T_{nn} are relaxation times which for the sake of simplicity we shall take as equals: $T_{mn}=T_{nn}=T$. z is the longitudinal propagation coordinate. ω_p is the pump frequency quasiresonant with the transition between levels 1 and 3, and ω_s is the signal frequency quasiresonant with the transition between levels 3 and 2; k_p and k_S are the wave numbers of the pump and the Stokes component, respectively. In Eq. (1), $\Delta_\perp$ is the transverse Laplace operator: $\Delta_\perp=\partial^2/\partial x^2+\partial^2/\partial y^2$, where x and y are transverse coordinates. $\varepsilon_p=\omega_p-\omega_{31}$ and $\varepsilon_s=\omega_s-\omega_{32}$ are detunings from one-photon resonances for the pump (ε_p) and the signal (ε_s) waves. ω_{31} and ω_{32} are the transitions frequencies (Fig. 1):

$$f_S(x,y,\varepsilon_s,\varepsilon_p)=\frac{2\pi\omega_s}{c}N\langle d\rangle\exp[-i(\omega_s t-k_S z)]\ ,$$

where N is the number of molecules in the unit volume, $\langle d\rangle$ is the quantum mechanically averaged value of the dipole momentum operator. This function will be calculated after solution of Eqs. (1) for the density-matrix elements. The solution of these equations for the off-diagonal density-matrix elements is represented in the form

$$\rho_{13}=A\exp[i(\omega_p t-k_p z)]\ ,$$
$$\rho_{32}=B\exp[-i(\omega_s t-k_S z)]\ , \qquad (2)$$
$$\rho_{12}=C\exp\{i[(\omega_p-\omega_s)t-(k_p-k_s)z]\}\ .$$

After substituting (2) into Eq. (1) and taking into account that

$$H_{mn}=\hbar\omega_{mm}\delta_{mn}-d_{mn}E(t,\mathbf{r})\quad(m,n=1,2,3)$$

(where δ_{mn} is the Kronecker's δ, d_{mn} is the matrix element of the dipole moment operator for the transition between levels m and n and $d_{nn}=0$), we obtain the following set of equations for the amplitudes A, B, and C of the off-diagonal density-matrix elements and for the population differences p_{13} and p_{32} defined as $p_{13}=\rho_{11}-\rho_{33}$, $p_{32}=\rho_{33}-\rho_{22}$:

$$\left[\frac{\partial}{\partial t}+\frac{1}{T}+i\varepsilon_p\right]A=-\frac{i}{2\hbar}(d_{13}p_{13}A_p+d_{23}CA_S)\ ,$$
$$\left[\frac{\partial}{\partial t}+\frac{1}{T}-i\varepsilon_s\right]B=\frac{i}{2\hbar}(d_{31}CA_p^*-d_{32}p_{32}A_S^*)\ ,$$
$$\left[\frac{\partial}{\partial t}+\frac{1}{T}+i\varepsilon\right]C=\frac{i}{2\hbar}(d_{13}BA_p-d_{32}AA_S^*)\ , \qquad (3)$$
$$\frac{\partial}{\partial t}p_{13}+\frac{p_{13}-p^0_{13}}{T}=\frac{i}{2\hbar}[2(d_{13}A_pA^*-d_{31}A_p^*A)+(d_{32}B^*A_S^*-d_{23}BA_S)]\ ,$$
$$\frac{\partial}{\partial t}p_{32}+\frac{p_{32}-p^0_{32}}{T}=\frac{i}{2\hbar}[(d_{31}AA_p^*-d_{13}A^*A_p)+2(d_{32}B^*A_S^*-d_{23}BA_S)]\ .$$

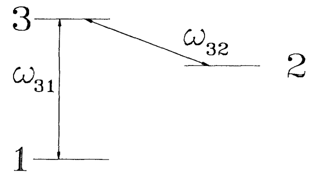

FIG. 1. Energy-level diagram for the three-level molecular system under consideration.

In the derivation of Eqs. (3), we ignore nonresonant terms and terms oscillating faster than ω_p or ω_s (the rotating-wave approximation). $d_{mn}=d^*_{nm}$ and single-photon transitions are assumed to be forbidden between levels 1 and 2: $d_{12}=0$. p^0_{13} and p^0_{32} are the equilibrium population differences in the absence of the laser fields. The detuning from the two-photon resonance ε is as follows:

$$\varepsilon=\omega_p-\omega_s-\omega_{21}=\varepsilon_p-\varepsilon_s .$$

Here we consider the steady-state solution of Eqs. (3) by making the population differences p_{13} and p_{32} as well as the amplitudes A, B, and C time independent, i.e., setting the time derivatives in (3) equal to zero. The case of a small signal and a strong pump wave is under consideration in the present paper and the influence of the small signal on the parameters of the material system has been ignored.

III. AMPLIFICATION OF A SMALL SIGNAL BY A COLLIMATED PUMP BEAM IN AN HOMOGENEOUSLY BROADENED MEDIUM

In this section small-signal gain is investigated in the field of a pump beam with spatial parameters, namely, transverse intensity distribution and the diameter of the beam may be assumed as the constants along the gain medium. The regime of high pump intensity when saturation and Stark splitting of the levels of the pump transition play an important role is under consideration.

Solution of the general system of Eqs. (3) for the steady-state case of homogeneously broadened molecular transitions gives the following expressions for the function $f_s(x,y,\varepsilon_p,\varepsilon_s)$ and population differences p_{13} and p_{32} (see also [18–20]):

$$\begin{aligned}
&f_s=f_1+if_2 ,\\
&f_1=R(Q_1x_s-Q_2),\quad f_2=R(Q_1+Q_2x_s) ,\\
&R=\frac{b_0V}{z_2(z_1+4V)},\quad b_0=\frac{2\pi\omega_s}{c}\frac{|d_{32}|^2Tp^0_{13}}{\hbar} ,\\
&Q_1=1+\frac{z_1z_2}{2(a_1^2+b_1^2)}[a_1(x_p+2Vx_s/z_2)\\
&\qquad -b_1(2V/z_2-1)] ,\\
&Q_2=\frac{z_1z_2}{2(a_1^2+b_1^2)}[b_1(x_p+2Vx_s/z_2)+a_1(2V/z_2-1)] ,\\
&a_1=z_1[x_s(z_2-V)-x_pz_2],\quad b_1=z_1(V+z_2) ,\\
&z_1=1+x_p^2, z_2=1+x_s^2, V=\left[\frac{|d_{13}||A_p|}{2\hbar}\right]^2T^2 ,\\
&p_{13}=p^0_{13}\frac{1+x_p^2}{1+x_p^2+4V} ,\\
&p_{32}=p^0_{13}\frac{2V}{1+x_p^2+4V} ,
\end{aligned}\tag{4}$$

where $x_s=\varepsilon_sT$ and $x_p=\varepsilon_pT$ are the normalized detunings for the signal and pump waves and it has been assumed that the molecules are in the ground state initially.

We assume Gaussian transverse distribution for the pump intensity:

$$|A_p|^2(r_\perp)=I_0\exp[-r_\perp^2/a_p^2] , \tag{5}$$

with I_0 being the peak intensity and a_p the radius of the pump beam, $r_\perp^2=x^2+y^2$. We use the Taylor power-series expansion of the function $f_s(r_\perp)$ about the beam center at $r_\perp=0$:

$$f_s(r_\perp,\varepsilon_s,\varepsilon_p)=f_s(r_\perp=0,\varepsilon_s,\varepsilon_p)+\frac{1}{2}\frac{\partial^2 f_s}{\partial r_\perp^2}\bigg|_{r_\perp=0}r_\perp^2 . \tag{6}$$

By substituting expression (6) into the last equation of (1) and assuming azimuthal symmetry, we represent the solution of the equation obtained in the factorized form

$$A_S(r_\perp,z)=\phi(x,z)\psi(y,z) , \tag{7}$$

with

$$\phi(x,z)=\phi(x)\exp[g_xz],\quad \psi(y,z)=\psi(y)\exp[g_yz] .$$

Then we obtain the following equation for the function $\phi(x)$:

$$\left[\frac{\partial^2}{\partial x^2}+(E_x-bx^2)\right]\phi(x)=0 , \tag{8}$$

where

$$E_x=-2ik_sq_x+\lambda_x ,$$

$$b=-k_s\frac{\partial^2}{\partial r_\perp^2}f_s\bigg|_{r_\perp=0} ,$$

with λ_x a free parameter.

For the function $\psi(x)$ we have an equation similar to Eq. (8) with the following change of notations: $E_x\to E_y$, $g_x\to g_y$, and $\lambda_x\to\lambda_y$, where

$$\lambda_x+\lambda_y=2k_sf_s(r_\perp=0,\varepsilon_s,\varepsilon_p) .$$

As follows from Eq. (7), the complex gain of the signal wave is equal to $G_s=g_x+g_y$.

Equation (8) formally coincides with the Schrödinger's equation for the wave function of a particle in an external field (see, for example, [21]) with the complex potential of the same dependence on the transverse coordinate ($r_\perp$) as for the function $f_s(r_\perp)$ [see Eq. (6)] which is proportional to the complex refractive index for the small signal in the plane-wave limit. The solutions of Eq. (8) are the Hermit polynomials $H_n(x)$, and we have for the function $\Phi(x,z)$:

$$\phi(x,z)=\exp[-\sqrt{b}x^2/2]\sum_{n=0}^{\infty}c_nH_n(xb^{1/4},g_x^{(n)})\times\exp(g_x^{(n)}z) , \tag{9}$$

where c_n are constants determined by the boundary conditions. The same expression (with $x\to y$ and $g_x\to g_y$) is valid for the $\psi(y,z)$. From the condition that the value of the $H_n(x)$ is limited when $x\to\infty$ we obtain the following expressions for the $g_x^{(n)}$ and $g_y^{(n)}$:

$$g_x^{(n)} = -i\frac{\lambda_x}{2k_s} + i(2n+1)\sqrt{b}\,/(2k_s)\ ,$$
$$g_y^{(m)} = -i\frac{\lambda_y}{2k_s} + (2m+1)\sqrt{b}\,/(2k_s)\ . \tag{10}$$

So we have for the signal complex gain,

$$G_s = G_{mn} = g_x^{(m)} + g_y^{(n)} = -if_s(r_\perp = 0, \varepsilon_s, \varepsilon_p) + \frac{i}{k_s}\sqrt{b}\,(m+n+1) \quad (m,n = 0,1,2,\dots)\ . \tag{11}$$

When $b \to 0$, we have from (11) the plane-wave limit $G_s^{(\mathrm{PW})}$ for the signal gain G_{mn},

$$G_{mn}(b \to 0) \to G_s^{(\mathrm{PW})} = -if_s(r_\perp = 0, \varepsilon_p, \varepsilon_s)\ .$$

As follows from the analysis of expression (11), the relation $\mathrm{Re}[G_{00}] \gg \mathrm{Re}[G_{mn}]$ $(m,n = 1,2,\dots)$ takes place and the real gain G of a small signal may be described by only the gain of the mode with $m = n = 0$:

$$G \cong \mathrm{Re}[G_{00}]\ .$$

The dependence of the gain G on the normalized signal frequency detuning x_s is presented in Figs. 2(a)–5(a) for various values of the pump normalized [see Eq. (4)] intensity V in the center of the beam $(r_\perp = 0)$ when an exact resonance for the pump wave takes place: $x_p = 0$.

To explain the offset, asymmetric deformation, and narrowing of the signal gain line represented in Figs. 2(a)–5(a), the functions of the real and the imaginary parts of the parameter b ($\mathrm{Re}[b]$ and $\mathrm{Im}[b]$) versus x_s are shown in Figs. 2(b)–5(b) at the same parameter values as in Figs. 2(a)–5(a), respectively.

The important role that the parameter b plays will be obvious from the following consideration: As follows from Eqs. (1) and (6), the pump-induced complex refractive index $n(r_\perp)$ of the medium is given by

$$n(r_\perp) = [f_s(r_\perp = 0) - br_\perp^2/(2k_s)]/k_s\ . \tag{12}$$

This would be a complex refractive index for the signal wave if no diffraction of the signal beam took place, i.e., when one could neglect the term $1/(2k)_s\Delta_\perp A_S \to 0$ in the left-hand side of the equation for A_S in (1). As is well known, the real part of the $n(r_\perp)$ describes a transverse distribution of the refractive index for the signal wave, when the imaginary one corresponds to the gain. So expression (12) describes an effective nonlinear waveguide with a gain (absorption) induced for the signal wave in the nonlinear medium by the strong pump beam. The parameter $\mathrm{Re}[b]$ is proportional to the square of the inverse value of the effective radius of this nonlinear waveguide. At the same time, as follows from Eq. (9), $\mathrm{Re}[b^{1/2}]$ is equal to the square of the inverse radius of the amplified signal beam. The wave guiding in the meaning of the total internal reflection is considered to take place when $\mathrm{Re}[b(x_s)] > 0$. In this case the refractive index from (12) has its maximum in the center of the beam and diminishes towards the periphery. In the opposite case, when $\mathrm{Re}[b] < 0$, there is no wave guiding at all and additional diffractive loss takes place because of a nonlinear defocusing of the signal beam.

In the spectral region where $\mathrm{Im}[b(x_s)] > 0$, the gain of the medium reaches its maximum value at the center of the beam and an active wave guiding (gain focusing) takes place. In the opposite case, of $\mathrm{Im}[b(x_s)] < 0$, we have gain defocusing for the signal wave.

The gain focusing $(\mathrm{Im}[b(x_s)] > 0)$ takes place for the all-frequency region of the signal at moderate values of the pump $(0 < V < 0.5)$, as can be seen in Figs. 2(b) and 3(b). At the same time a nonlinear wave guiding takes place in the region where $\mathrm{Re}[b(x_s)] > 0$ and additional diffraction losses occur at $\mathrm{Re}[b(x_s)] < 0$.

The mutual action of these gain and nonlinear waveguides leads to the asymmetry of the signal gain line and to the appearance of an absorption in the spectral region for the sufficiently large (according to the absolute values) negative x_s. The increase of the pump intensity ($V > 0.5$ and for the values of the other parameters applied in Figs. 2–5) leads to the Stark splitting of the levels

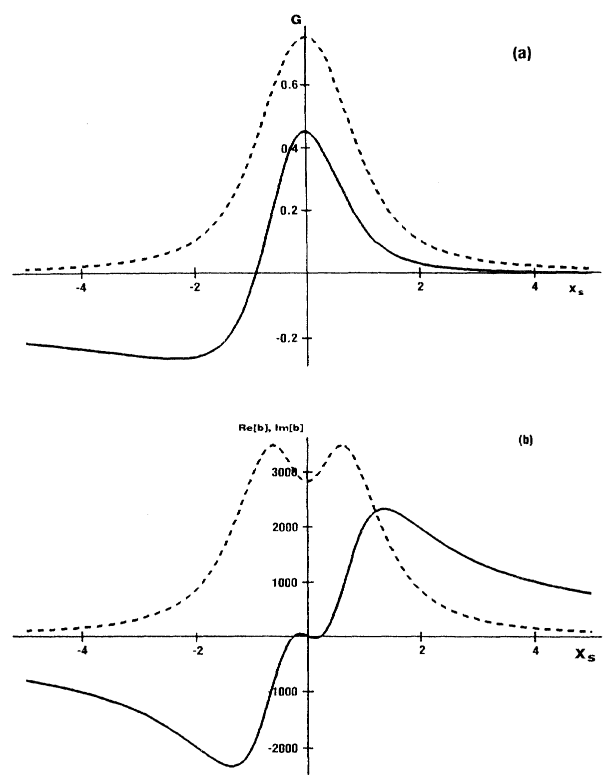

FIG. 2. (a) Dependence of the small signal gain on the signal normalized frequency detuning x_s in the case of a homogeneously broadened pump transition when an exact resonance for the pump wave takes place: $x_p = 0$. The values of the parameters are as follows: $V = 0.25$, $a = 0.1$ cm, $b_0 = 5$ cm^{-1}, $k_s = 20\pi$ cm^{-1}. The dashed curve corresponds to the plane-wave pump limit. (b) Dependences of the real (solid line) and imaginary (dashed line) parts of the parameter b on the frequency detuning x_s at the same parameter values as in 2(a).

of the pump transition and to the appearance of a region of the gain defocusing ($\mathrm{Im}[b(x_s)]<0$) in the central region of the gain line [see Fig. 4(b), where the value $V=1$]. The same arguments used above can be applied to explain the peculiarities of the function of the gain versus x_s in Figs. 4(a) and 5(a) by considering the frequency behavior of the functions $\mathrm{Re}[b(x_s)]$ and $\mathrm{Im}[b(x_s)]$ depicted in Figs. 4(b) and 5(b).

For the real gain $G=\mathrm{Re}[G_{00}]$, we have the following expression from Eq. (11):

$$G=G_s^{(\mathrm{PW})}-\mathrm{Im}[b^{1/2}]/k_s \ .$$

The term proportional to the $\mathrm{Im}[b^{1/2}]$ in this expression describes the decrease of the gain because of the diffraction losses. The gain $G(x_s)$ reaches its maximum value at the signal frequency $x_s=x_s^{(*)}$ at which the function

$$\mathrm{Im}[b^{1/2}(x_s)]=\{[(\mathrm{Re}[b])^2+(\mathrm{Im}[b])^2]^{1/2}-\mathrm{Re}[b]^{1/2}\}/\sqrt{2} \tag{13}$$

takes on its minimum value. Note that $\mathrm{Im}[b^{1/2}]\geq 0$ because of the presence of the diffraction losses. As follows from (13), this minimum value has been achieved when the function $(\mathrm{Im}[b])^2$ takes on its minimum value as well. The larger the pump intensity, the smaller the minimum value $\min\{(\mathrm{Im}[b])^2\}$ and the larger the maximum value $\max\{G\}$ of the gain (see Figs. 2 and 3).

This is true in the region of the moderate values of the pump intensity where no Stark splitting of the levels of the pump transition takes place. For sufficiently large values of the pump intensity, $\mathrm{Im}[b(x_s)]$ becomes negative in some region of the x_s. In this region of detuning x_s, we have a gain defocusing instead of a gain focusing, and the amplification of the signal in the center of the pump beam is smaller than in the periphery. The reason for such a behavior is an appearance of a nonlinear detuning from the resonance of the pump and signal waves due to the Stark splitting of the levels of the pump transition and the saturation of the gain as well. In this situation the minimum value of the function $\{\mathrm{Im}[b(x_s)]\}^2$ is equal to zero. This minimum value has been achieved at the signal frequency $x_s=x_s^{(*)}(V)$ [see Figs. 4(b) and 5(b)]. In this case, in accordance with Eqs. (13) and (11), $\mathrm{Im}[b^{1/2}(x_s^{(*)})]=0$, and the signal gain G takes on its maximum value which in the case under consideration and in the limits of the approximation applied [see Eq. (8)] coincides with the gain in the plane-wave limit. It should be noted that there are two values of x_s ($x_s^{(*)}$ and $x_s^{(1)}$) at which the function $\mathrm{Im}[b(x_s)]$ takes on the zero value. Only at the one of them denoted by the $x_s^{(*)}$ ($x_s^{(*)}<0$) in Figs. 4 and 5 does $\mathrm{Re}[b(x_s^{(*)})]>0$, and nonlinear focusing takes place which leads to the enhancement of the signal gain. At the second one ($x_s=x_s^{(1)}$), $\mathrm{Re}[b(x_s^{(1)})]<0$, and the nonlinear defocusing of the signal wave causes decrease of the gain. At the frequency $x_s^{(*)}$ the pump-induced active waveguide changes its character from gain focusing at $x_s<x_s^{(*)}$ to gain defocusing at $x_s^{(*)}>x_s<x_s^{(1)}$ [see Eq. (12) and Figs. 4(b) and 5(b)]. The asymmetry of the nonlinear wave guiding and

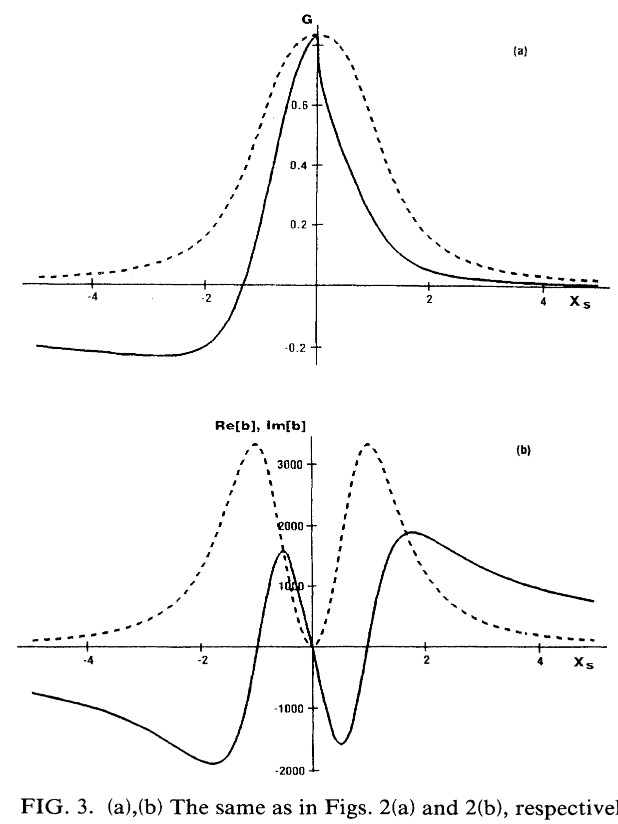

FIG. 3. (a),(b) The same as in Figs. 2(a) and 2(b), respectively, with $V=0.5$.

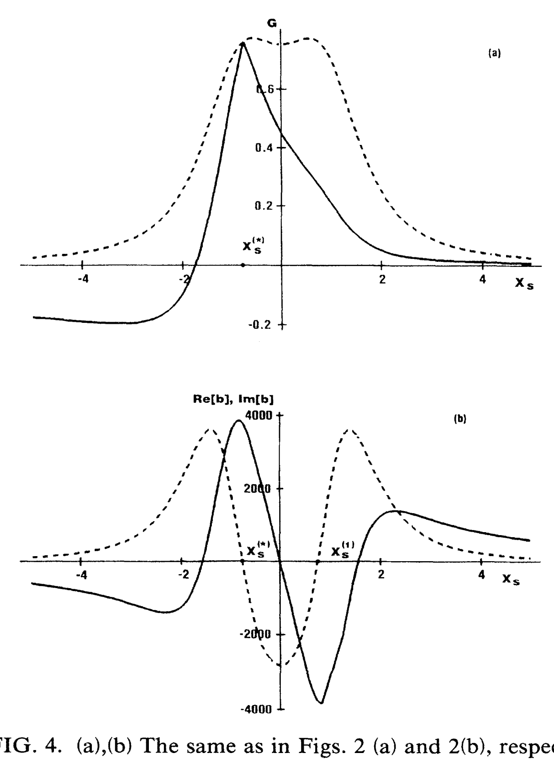

FIG. 4. (a),(b) The same as in Figs. 2 (a) and 2(b), respectively, with $V=1$.

the gain focusing of the signal wave in its frequency domain lead to an asymmetric deformation, a shifting, and a narrowing of the gain line shape. This narrowing may be significant at sufficiently high values of the pump intensity. In analogy with the effects of gain or nonlinear (dispersion) focusing (see, for example, [2–4]), the effect obtained in this paper may be termed signal spectrum focusing. To demonstrate this effect, we show in Fig. 5(a) the signal gain line at sufficiently high pump intensity.

Note that the function $\mathrm{Re}[b(x_s)]$ takes on its maximum value and the diameter of the nonlinear waveguide reaches its minimum value at the signal frequency which is equal to the $x_s^{(*)}$. The numerical estimations show that at $x_s = x_s^{(*)}$ in Figs. 4 and 5, the diameter of the amplified signal is much smaller than that of the pump beam.

IV. MOLECULAR SYSTEM WITH DOPPLER-BROADENED PUMP TRANSITION

In order to consider the influence of Doppler broadening on the gain line shape of the small signal, let us denote by v the velocity component of molecules along the propagation direction of the pump and signal waves. In a coordinate system in which the ensemble of molecules is at rest due to the Doppler effect, we must replace the frequencies of the waves ω_p and ω_s by

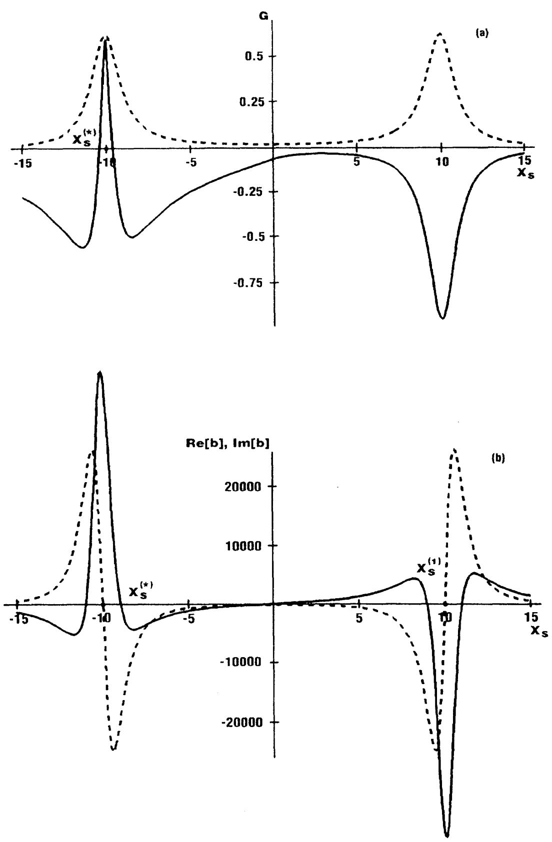

FIG. 5. (a),(b) The same as in Figs. 2(a) and 2(b), respectively, with $V = 100$.

$$\omega_p \to \Omega_p = \omega_p - k_p v, \quad \omega_s \to \Omega_s - k_s v .$$

Let $W(v)$ be the velocity distribution of molecules:

$$W(v) = \frac{1}{\pi^{1/2} v_T} \exp[-(v/v_T)^2] ,$$

where v_T is the thermal velocity $v_T = (2kT/M)^{1/2}$, with M being the mass of the active molecule.

In order to calculate the induced polarization $\langle d \rangle_{\mathrm{Dp}}$ of the medium in the case of a Doppler-broadened transition, one has to average the function $\langle d(v) \rangle$ over the velocity distribution $W(v)$:

$$\langle d \rangle_{\mathrm{Dp}} = \int_{-\infty}^{\infty} \langle d(v) \rangle W(v) dv . \tag{14}$$

Here we assume that only the pump transition is Doppler broadened. So let the Doppler effect of the FIR signal frequency ω_s be negligible compared to the pressure-broadened linewidth. Thus, the following conditions have been assumed:

$$\gamma = 2/T \gg k_s v_T \quad \text{and} \quad \gamma \ll k_p v_T .$$

Therefore, in the velocity integration in (14) we may neglect the $k_s v$ velocity dependence by putting $\Omega_s = \omega_s$, but $\Omega_p = \omega_p - k_p v$. After integration, we obtain the following expression for the function f_s [see Eq. (4)] for the case of a Doppler-broadened pump and homogeneously broadened signal transitions:

$$\begin{aligned} f_s(r_\perp, x_s) &= i d_0 \frac{V[1+(q+1)/2 + i x_s]}{q[V + (1+ix_s)(1+q+ix_s)]} \\ &\quad \times \exp\{-[x_p \gamma/(k_p v_T)]^2\} , \\ d_0 &= \frac{4\pi\omega_s |d_{23}|^2 p_{13}^0}{c\hbar k_p v_T/\sqrt{\pi}}, \quad q = (1+4V)^{1/2} , \\ V &= \left[\frac{|d_{13}||A_p|}{2\hbar} \right]^2 T^2 . \end{aligned} \tag{15}$$

Assuming that the pump intensity is of Gaussian transverse distribution (5), and after substituting the expression (15) for the function $f_s(r_\perp, x_s, x_p)$ into Eq. (11), we obtain the complex gain for the small signal. The real part of it is the signal gain for the case of a three-level molecular system with Doppler-broadened pump transition. In Fig. 6 the signal gain versus signal frequency detuning x_s is depicted for various values of the pump peak intensity when an exact resonance for the pump wave ($x_p = 0$) takes place. As can be seen from Fig. 6, an offset of the signal gain line (solid curves) with a region of absorption takes place in the case of a transversely nonuniform pump beam. For comparison, the functions of the corresponding gain versus x_s are presented in Fig. 6 for the plane wave limit. As one can see, the increase of the pump intensity leads to a broadening of the gain line. It is interesting to note that the gain-line maximum has been shifted to the region of larger signal frequencies in the case of relatively small pump intensities. The increase of the pump intensity leads to the shift into the opposite direction, i.e., to the region of smaller signal frequencies.

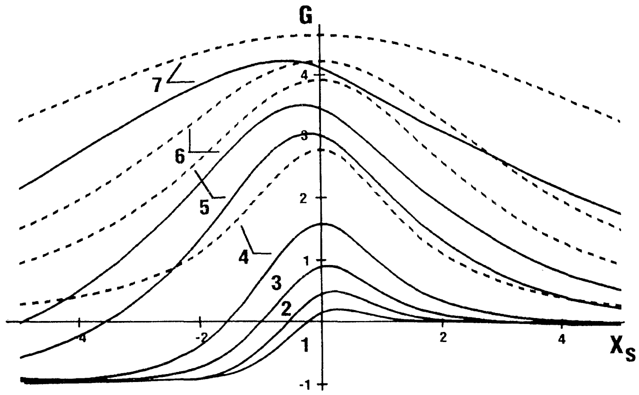

FIG. 6. Dependence of the small signal gain on the signal frequency detuning x_s in a molecular system with a Doppler-broadened pump transition for some values of the normalized pump intensity V: 1.0$-V=0.2$, 2.0$-V=0.3$, 3.0$-V=0.5$, 4.0$-V=1$, 5.0$-V=5$, 6.0$-V=10$, and 7.0$-V=50$ with the following parameters applied: $d_0=10$ cm^{-1}, $k_s=20\pi$ cm^{-1}, $a=0.1$ cm. The dashed curves correspond to the plane-wave pump limit.

V. CONCLUSION

To summarize, the results obtained by investigating a small-signal-gain line in a three-level vibrational-rotational Λ-type molecular system are presented. The saturation and dynamic Stark effects on the pump transition have been taken into account. The cases of homogeneous and Doppler-broadened pump transitions have been considered, when homogeneous broadening for the signal transition in both cases has been assumed. A significant offset, asymmetric deformation, and narrowing of the gain line, termed signal spectrum focusing, as well as the appearance of an absorption region in the gain line, have been obtained. An offset of the gain-line maximum to the lower signal frequencies has been obtained in the case of a homogeneously broadened pump transition. An offset in both directions, i.e., to the higher signal frequencies and to the lower ones, depending on the pump-beam peak intensity, has been obtained for the gain line in the case of an inhomogeneously broadened pump transition. These peculiarities have been explained as a result of the influence of active (gain) and nonlinear (dispersion) waveguides induced by the strong transversely nonuniform pump beam in the molecular system when the saturation and dynamic Stark effects of the pump transition have been taken into account.

The results of the present analysis should be taken into account when spectroscopic, Dicke's superradiance investigations, or signal amplification are performed in a gain medium, e.g., gaseous far-infrared or Raman laser, and, in particular, when a pump beam as a spatial solitary wave has been applied. An important application of the effect of spectrum focusing is narrow-band filtering with amplification.

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