

## Small-signal spectrum focusing using a strong nonuniform pumping beam

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Significant offset and narrowing of the small-signal-gain line, i.e., spectrum focusing, in a three-level molecular system at the regime of the saturation and Stark (Autler-Townes) splitting of the pumping transition in the field of a strong collimated pump beam have been obtained and explained.

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Focusing or collimation of the pumping laser beam is usually used when small-signal amplification is performed. The nonuniform intensity distribution of the pump beam yields a corresponding nonuniform transverse distribution for the gain and refractive index of the medium. The problem of propagation of the electromagnetic wave in the presence of a transverse quadratic gain or (and) dispersion variation was analyzed in [1–4]. The transverse distribution of the intensity of the pump beam was approximated by the square of the hyperbolic secant function in [5], in which stimulated Raman scattering (SRS) was investigated. A number of authors have dealt with the pump-beam focusing [6–8]. The SRS was investigated by taking into account the depletion of the pump beam [9] and by including Stokes–anti-Stokes interaction in [10]. All previous investigations dealt with either two-level media or were performed without taking into account saturation of the transitions and Stark (Autler-Townes) splitting of the levels.

This paper addresses an important practical problem for investigating the small-signal-gain line shape in a three-level vibrational-rotational molecular system in the field of a collimated strong laser pump beam when the saturation of the pumping transition and dynamic Stark (Autler-Townes) effect play an important role.

For infrared–far-infrared double-resonance spectroscopic [11] or Dicke superradiance investigations [12], 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 the rotational levels of the vibrationally excited manifold. Thus, the wavelengths of the pump and 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 but the diffraction of the amplified signal must be taken into account. We show that the gain line shape for a small signal obtained for the collimated transversely nonuniform pump beam in the  $\Lambda$ -type three-level molecular system, taking into account the transition saturation and dynamic Stark effect on the pump transition, can significantly differ from that for the plane uniform pump wave. Offset and significant narrowing of the gain line have been obtained and interpreted as a result of the influence of the active

waveguide induced by the pump beam in the nonlinear medium combined with the Stark splitting and saturation of the levels of the pump transition. This active waveguide is the result of the nonuniform transverse intensity distribution of the pump beam which induces the corresponding spatial distribution of the gain and refractive index for the amplified signal. The nonlinear wave guiding occurs only for some frequency region of the signal. Spectral narrowing of the gain line in the former frequency region takes place with increasing pump intensity as a result of the decrease of the effective diameter of the pump-induced nonlinear waveguide.

Our results are presented in Figs. 1 and 2 where the small-signal-gain line shapes are plotted for a plane-wave pump and collimated pump beam whose intensity equals that of the plane wave. Compared with the plane waves, it is apparent that offset, deformation, and narrowing of the gain line take place for the collimated pump beam. The higher the pump intensity, the larger the difference.

A simple derivation of these results is as follows. Consider the interaction between a weak signal wave of complex amplitude  $A_s$  and frequency  $\omega_s$  and a strong pump beam of amplitude  $A_p$  and frequency  $\omega_p$  in a three-level molecular system (see inset in Fig. 1). The scalar wave equation governing the signal wave amplitude in the stationary regime of interaction in the paraxial approximation has the form

$$\left[ \frac{\partial}{\partial z} + \frac{i}{2k_s} \Delta_{\perp} \right] A_s = -if_s(r_{\perp}, \epsilon_s, \epsilon_p, |A_p|^2) A_s, \quad (1)$$

where  $z$  is the longitudinal propagation coordinate,  $k_s$  is the wave vector of the signal, and  $\Delta_{\perp}$  is the transverse Laplace operator which, for simplicity, is assumed to be one dimensional, i.e.,  $\Delta_{\perp} = \partial^2 / \partial x^2$  with  $x = r_{\perp}$  being the transverse coordinate. (Generalization to the two-dimensional case is easily done.)  $\epsilon_p = \omega_p - \omega_{31}$  and  $\epsilon_s = \omega_s - \omega_{32}$  are the detunings from one-photon resonances for the pump ( $\epsilon_p$ ) and the signal ( $\epsilon_s$ ) waves.  $\omega_{31}$  and  $\omega_{32}$  are the transition frequencies (see Fig. 1).

The expression for  $f_s$  at the steady-state regime of interaction for a weak signal wave taking into account the transition saturation and the Stark splitting of the levels of the pump transition of the three-level system (Fig. 1) has the following form (see also [13,14]):

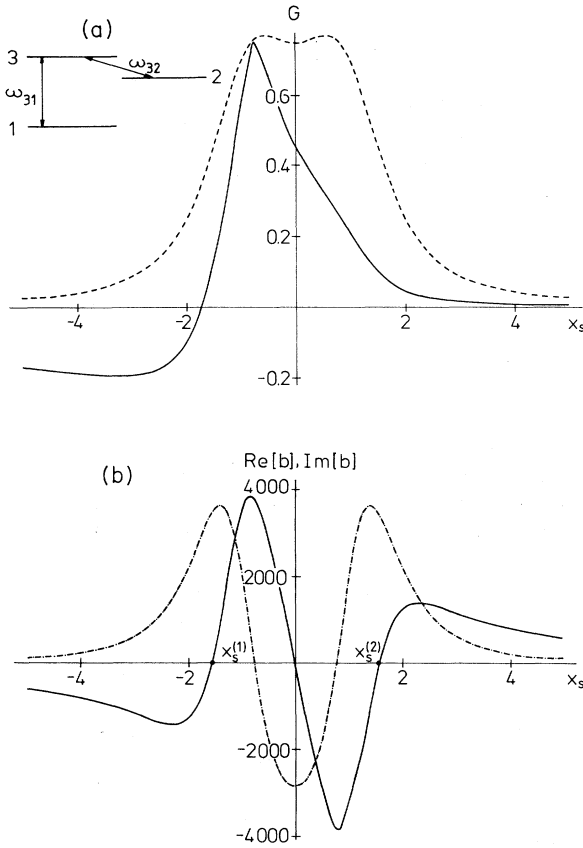


FIG. 1 (a) Dependence of signal gain  $G$  on signal frequency detuning  $x_s$  at  $V(x=0)=1$ ,  $b_0=5 \text{ cm}^{-1}$ ,  $a=10^{-1} \text{ (cm)}$ ,  $k_s=20\pi \text{ (cm}^{-1}\text{)}$ , and  $x_p=0$ . The dotted curve depicts the plane-wave limit of  $G$ . Inset: scheme of the three-level system. (b) Dependence of the  $\text{Re}[b]$  (solid curve) on signal frequency detuning  $x_s$ , at the same values of parameters as in (a). The dotted line describes  $\text{Im}[b]$ .

$$f_s = f_1 + if_2,$$

$$f_1 = R(Q_1 x_s - Q_2), \quad f_2 = R(Q_1 + Q_2 x_s)$$

$$R = \frac{b_0 V}{z_2(z_1 + 4V)}, \quad b_0 = 2\pi\omega_s \frac{|d_{32}^2| T n_{13}^0}{c\hbar},$$

$$Q_1 = 1 + \frac{z_1 z_2}{2(a_1^2 + b_1^2)} [a_1(x_p + 2Vx_s/z_2) - b_1(2V/z_2 - 1)],$$

(2)

$$Q_2 = \frac{z_1 z_2}{2(a_1^2 + b_1^2)} [b_1(x_p + 2Vx_s/z_2) + a_1(2V/z_2 - 1)],$$

$$a_1 = z_1[x_s(z_2 - V) - x_p z_2], \quad b_1 = z_1(V + z_2),$$

$$z_1 = 1 + x_p^2, \quad z_2 = 1 + x_s^2, \quad V = \left[ \frac{|d_{13}| |A_p|}{2\hbar} \right]^2 T^2.$$

Here  $x_x = \varepsilon_s T$ ,  $x_p = \varepsilon_p T$ , and  $d_{13}$  and  $d_{32}$  are the matrix elements of the electric dipole moment for the corresponding transitions in the three-level system of Fig. 1.

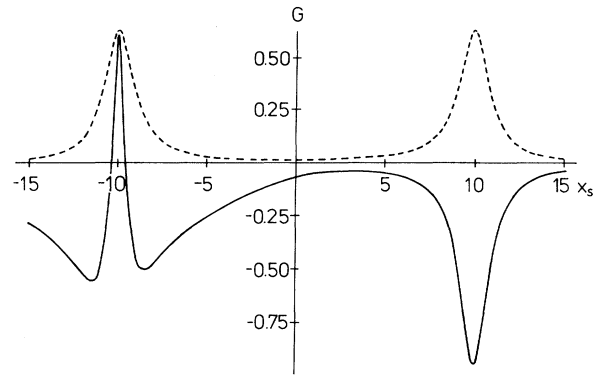


FIG. 2. Signal gain  $G$  vs detuning  $x_s$  at  $V(x=0)=100$ ; other parameters as in Fig. 1.

$n_{13}^0$  is the initial difference of the populations for the pumping transition. For simplicity the longitudinal and transverse relaxation times of this system are taken to be the same and are equal to  $T$ .

We assume a Gaussian transverse distribution for the pump intensity:

$$|A_p|^2(x) = I_0 \exp[-(x/a)^2],$$

with  $I_0$  being the peak intensity and  $a$  the radius of the pump beam. We use the expansion of function  $f_s(x)$  in a Taylor-power series of the transverse coordinates  $x$  about the beam center at  $x=0$ :

$$f_s(x, \varepsilon_s, \varepsilon_p) = f_s(x=0, \varepsilon_s, \varepsilon_p) + \frac{1}{2} \frac{\partial^2 f_s}{\partial x^2} \Big|_{x=0} x^2. \quad (3)$$

The solution of the equation obtained by substituting expression (3) into (1) is as follows:

$$A(x, z) = \exp[-\sqrt{b} x^2 / 2] \sum_{n=0}^{\infty} c_n H_n(xb^{1/4}) \exp[G_n z]. \quad (4)$$

Here  $H_n(x)$  are the Hermite polynomials and  $c_n$  are constants determined by the boundary condition:  $A(x, z=0) = A_0(x)$  with  $A_0(x)$  describing the amplitude of the signal at the entrance ( $z=0$ ) of the medium:

$$b = -k_s \frac{\partial^2 f_s}{\partial x^2} \Big|_{x=0},$$

$$G_n = -if_s(x=0, \varepsilon_s, \varepsilon_p) + i(2n+1)\sqrt{b}/(2k_s) \quad (5)$$

$$(n=0, 1, 2, \dots).$$

When  $b \rightarrow 0$  we have from (5) the plane-wave limit for the signal gain  $\text{Re}[G_n]$  (for details see, for example, [13,14]).

Analysis of expression (5) shows that the relation  $\text{Re}[G_0] \gg \text{Re}[G_n]$  ( $n=1, 2, \dots$ ), is fulfilled for the Gaussian pump beam, and the small signal gain  $G$  is determined by the gain mode of the lowest order, i.e.,

$$G \cong \text{Re}[G_0].$$

The dependence of gain  $G$  on signal frequency detuning  $x_s$  is presented in Fig. 1(a) at a fixed value of  $x_p$  ( $x_p=0$ ) and fixed values of the pump peak intensity.

To explain the offset, deformation, and narrowing of the signal-gain line in Fig. 1(a), the real and imaginary parts of parameter  $b$  are depicted versus  $x_s$  in Fig. 1(b) with the same values of parameters as in Fig. 1(a). It follows from Eqs. (1)–(3) that the pump-induced complex dielectric constant  $\varepsilon(x)$  of the medium is given by

$$\varepsilon(x) = 2[f_s(x=0) - bx^2/(2k_s)]/k_s. \quad (6)$$

Thus  $\text{Re}[b]$  is proportional to the square of the inverse value of the effective radius of the pump-induced nonlinear waveguide. At the same time, as it follows from Eq. (4),  $\text{Re}[b^{1/2}]$  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 have taken place when  $\text{Re}[b(x_s)] > 0$ . In this case the refractive index has its maximum in the center of the beam and diminishes towards the periphery. In the opposite case, when  $\text{Re}[b] < 0$ , there is no wave guiding at all and additional diffractive loss takes place. Figure 1(b) shows function  $\text{Re}[b(x_s)] > 0$  for  $x_s^{(1)} < x_s < 0$  and  $\text{Re}[b(x_s)] < 0$  for  $0 < x_s < x_s^{(2)}$  in the region of the maximum values of the gain in the plane-wave limit [see Fig. 1(b)].

It follows from this that the gain of the small signal must be enhanced for  $x_s < 0$  and must be suppressed for the signal frequencies  $x_s > 0$ . These features of the active waveguide make it possible to explain the peculiarities of the gain dependence on  $x_s$  in Fig. 1(a).

The term proportional to  $b^{1/2}$  in Eq. (5) describes the decrease of the gain because of the diffraction losses. The signal gain  $\text{Re}[G_0]$  has its maximum value at the signal frequency  $x_s = x_s^{(*)}$  at which the function

$$\text{Im}[b^{1/2}(x_s)] = \{[\text{Re}(b)^2 + \text{Im}(b)^2]^{1/2} - \text{Re}(b)\}^{1/2}/\sqrt{2} \quad (7)$$

takes on its minimum value. Note that  $\text{Im}[b^{1/2}] \geq 0$  because of the presence of the diffraction losses. The function  $\text{Im}[b(x_s)]$  takes on the value equal to zero at the signal frequency  $x_s = x_s^{(*)}$ —as is depicted in Fig. 1(b). In this case, in accordance with Eqs. (7) and (5),  $\text{Im}[b^{1/2}] = 0$  and the signal gain  $\text{Re}[G_0]$  takes on its maximum value which, in the case under consideration, coincides with the gain in the plane-wave limit. It should be noted that this coincidence only takes place if the pump intensity is high.

In our case the frequency  $x_s^{(*)}$  is one of the two frequencies of the signal wave at which the pump-induced waveguide changes its character from being gain focusing at  $x_s < x_s^{(*)}$  to being gain defocusing at  $x_s > x_s^{(*)}$  [see Eq. (6) and Fig. 1(b)]. At the same time  $\text{Re}[b(x_s)]$  takes on its maximum value and the diameter of the nonlinear waveguide reaches its minimum value at the signal frequency which equals  $x_s^{(*)}$ . The numerical estimations show that in this case the diameter of the amplified signal is much smaller than that of the pump beam. In analogy with the effects of gain or dispersion focusing (see, for example, [3,4]) the effect obtained in this paper may be termed signal spectrum focusing. To demonstrate this effect we show (Fig. 2) the signal-gain line at sufficiently high pump intensity.

It must be noted that the dynamic Stark effect and saturation of the levels of the pump transition play an important role in the effect of spectrum focusing described in the present paper. This effect differs substantially from insignificant offset and deformation of the Raman gain line obtained in [4,5,15] where large detunings from one-photon resonances for both pump and signal (Stokes) waves have been assumed and no dynamic Stark and saturation effects have been taken into account. When an offset of the Raman gain line center to the higher signal (Stokes) frequencies has been obtained in these papers, a significant offset of the signal-gain line to the lower signal frequency region has been obtained in the present paper.

To summarize, the results obtained by investigating a small-signal-gain line in a three-level vibrational-rotational molecular system are presented. The saturation and the dynamic Stark effect of the pump transition have been taken into account. Significant offset, deformation, and narrowing of the gain line, viz., the effect of the spectrum focusing of the amplified signal, have been obtained. These peculiarities are explained as a result of the competition between the gain of the medium and the active wave guiding induced in the nonlinear gain medium by the strong pump beam.

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 lasers. An important application of the effect of the spectrum focusing is for narrow-band filtering with amplification.

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