Spectroscopy within the radiative linewidth

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A method is proposed for resolving close spectral lines when the transition frequency difference is less than the radiative line width Γ. It is shown that the fine structure can be studied when the difference δ is less by a factor of a few hundreds than the homogeneous (radiative or collisional) width.

INTRODUCTION

The methods of nonlinear spectroscopy can now be used to isolate spectral lines against the background of a relatively broad Doppler profile when they are not masked by homogeneous broadening, i.e., radiative or collisional (see, for example, [1]). This resolution of spectral lines is based on the hole burning of the Doppler line which leads to the appearance of intensity resonances in the emission of gas lasers. These resonances, the centers of which fix the transition frequencies, have widths of the order of the homogeneous line width. It follows that when the transition frequency difference δ is less than the homogeneous line width, the separation of the individual spectral components becomes impossible.

In this paper, we propose a method which can be used to find the shift δ between two spectral components of a line even when δ is much less than the homogeneous width. The method is based on the competition between spatial and frequency hole burning of the medium in a ring gas laser.

The dependence of the region of existence of very different operating conditions of the ring laser (standing and traveling waves) on the frequency difference δ can, in principle, be used to detect the presence of fine structure when δ is smaller by a factor of several hundreds than the homogeneous (radiative or collisional) line width. This was previously noted in [2].

THEORY

1. Consider the possible separation of two close spectral components with frequencies ω_{ac} and ω_{bc} corresponding to the b → a and c → a transitions (Fig. 1). The material equations for the laser, i.e., the equations for the elements of the density matrix ρ_{ij}, are

$$\begin{aligned}
\left(\frac{\partial}{\partial t}+v\frac{\partial}{\partial x}\right)\rho_{ab}&=-i\omega\rho_{ab}-\gamma_{ab}\rho_{ab}+iV_{ab}(\rho_{ac}-\rho_{bb}),\\
\left(\frac{\partial}{\partial t}+v\frac{\partial}{\partial x}\right)\rho_{bb}&=-\Gamma_b(\rho_{bb}-\rho_{bb}^{(0)})-iV_{ab}(\rho_{ab}-\rho_{ba}),\\
\left(\frac{\partial}{\partial t}+v\frac{\partial}{\partial x}\right)\rho_{aa}&=-\Gamma_a(\rho_{aa}-\rho_{aa}^{(0)})+iV_{ab}(\rho_{ab}-\rho_{ba})+iV_{ac}(\rho_{ac}-\rho_{ca}),\\
\left(\frac{\partial}{\partial t}+v\frac{\partial}{\partial x}\right)\rho_{ac}&=-i\Omega\rho_{ac}-\gamma_{ac}\rho_{ac}+iV_{ac}(\rho_{aa}-\rho_{cc}),\\
\left(\frac{\partial}{\partial t}+v\frac{\partial}{\partial x}\right)\rho_{cc}&=-\Gamma_c(\rho_{cc}-\rho_{cc}^{(0)})-iV_{ac}(\rho_{ac}-\rho_{ca}),
\end{aligned}\tag{1}$$

where ω and Ω are the transition frequencies for a → b and a → c, $\hbar V_{ab} = -\mu_{ab}E$ and $\hbar V_{ac} = -\mu_{ac}E$ are the energies of these two transitions in the laser field E, μ_{ab} and μ_{ac} are the matrix elements of the dipole moments of the transition, γ_{ab}, γ_{ac} and Γ_a, Γ_b, Γ_c are the longitudinal and transverse relaxation constants, $\rho_{aa}^{(0)}$, $\rho_{bb}^{(0)}$, $\rho_{cc}^{(0)}$ are the level populations in the absence of the field, and x and v are the coordinates and velocities of the molecules.

We shall suppose that

$$\gamma_{ab}=\gamma_{ac}=\gamma,\quad \Gamma_a=\Gamma_b=\Gamma_c=\Gamma,\quad V_{ab}=V_{ac}=V,\quad \rho_{bb}^{(0)}=\rho_{cc}^{(0)}.\tag{2}$$

Physically, this means that the profiles of the spontaneous lines corresponding to the a → b and a → c transitions are frequency-shifted, but all the other characteristics for the b → a and c → a transitions are the same. We shall suppose, moreover, that the frequency difference $(\Omega - \omega)$ is small in comparison with the homogeneous width γ of the lines. Thus, the spontaneous line profiles overlap even in the case of fixed atoms and, consequently, according to the criteria adopted in optics, the transitions cannot be resolved.

To solve (1) for a strong laser field E, we can use the standard procedure for resolving the elements of the density matrix into a Fourier series, and obtain an expression for the nonlinear correction to the polarization of the medium in third-order perturbation theory to within terms of the order of $(\gamma/ku)^2$ inclusive (k is the wave vector and u the thermal velocity of the molecules). However, this is a very laborious procedure. It can be avoided if we recall that, in a certain definite approximation, the equations given by (2) can be reduced to the equations describing a medium as a mixture of two isotopes of a gas. The solution of these equations is well known (see, for example, [3]). We shall now prove this.

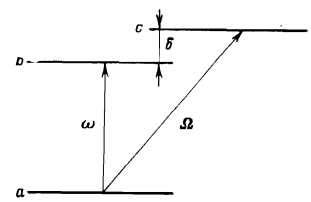

FIG. 1

2. The polarization of the medium is given by the sum of the nondiagonal elements of the density matrix:

$$P(t,x)\sim\int dv[\rho_{ab}+\rho_{ac}+\text{c.c.}].\tag{3}$$

To determine the quantity $\rho_{ab} + \rho_{ac}$ in (3), we use (1) to show that

$$(\partial/\partial t+v\partial/\partial x)X=-i\omega_0X+{}^1/_2i\delta\bar{X}-\gamma X+iVU,\tag{4}$$

$$(\partial/\partial t+v\partial/\partial x)U=-\Gamma(U-U^0)+3iV(X-X^*),\tag{5}$$

$$(\partial/\partial t+v\partial/\partial x)\bar{X}=-i\omega_0\bar{X}+{}^1/_2i\delta X-\gamma\bar{X}-iV\bar{Z},\tag{6}$$

$$(\partial/\partial t+v\partial/\partial x)\bar{Z}=-\Gamma\bar{Z}-iV(\bar{X}-X^*),\tag{7}$$

where

$$X=\rho_{ab}+\rho_{ac},\ U=2\rho_{aa}-(\rho_{bb}+\rho_{cc}),\tag{8}$$

We shall be interested in the solution of (4)–(7) to within terms of the order of E^3, inclusive. It is readily seen that, in this approximation, $\tilde{Z} = 0$. Hence, the equations for the density matrix assume the form

$$U^{(0)}=2\rho_{aa}^{(0)}-(\rho_{bb}^{(0)}+\rho_{cc}^{(0)}),\quad \delta=\Omega-\omega,$$
$$\omega_0=(\omega+\Omega)/2,\ X=\rho_{ab}-\rho_{ac},\ Z=\rho_{bb}-\rho_{cc},\qquad (9)$$
$$Z^{(0)}=\rho_{bb}^{(0)}-\rho_{cc}^{(0)}.$$

$$(\partial/\partial t+v\partial/\partial x)X=-i\omega_0X+{}^1/_2i\delta\bar{X}-\gamma X+iVU,$$
$$(\partial/\partial t+v\partial/\partial x)\bar{X}=-i\omega_0\bar{X}+{}^1/_2i\delta X-\gamma\bar{X},\qquad (10)$$
$$(\partial/\partial t+v\partial/\partial x)U=-\Gamma(U-U^0)+3iV(X-X^*),$$

The equations for the density matrix describing the state of a gas with a 1 : 1 isotopic composition can be reduced to a form analogous to (10) (Fig. 2). In the cubic approximation in E we find, in this way, that

$$\left(\frac{\partial}{\partial t}+v\frac{\partial}{\partial x}\right)X=-i\omega_0X+\frac{1}{2}i\delta\bar{X}-\gamma X+iVU,$$
$$\left(\frac{\partial}{\partial t}+v\frac{\partial}{\partial x}\right)\bar{X}=-i\omega_0\bar{X}+\frac{1}{2}i\delta X-\gamma\bar{X},\qquad (11)$$
$$\left(\frac{\partial}{\partial t}+v\frac{\partial}{\partial x}\right)U=-\Gamma(U-U^{(0)})+2iV(X-X^*),$$

where

$$U^{(0)}=(\rho_{aa}^{(0)}+\rho_{a'a'}^{(0)})-(\rho_{bb}^{(0)}+\rho_{cc}^{(0)}),\qquad (12)$$

in which

$$X=\rho_{ab}+\rho_{a'c},\ \bar{X}=\rho_{ab}-\rho_{a'c},$$
$$U=(\rho_{aa}+\rho_{a'a'})-(\rho_{bb}+\rho_{cc}).\qquad (13)$$

In deriving (11), we assume that $\rho_{aa}^{(0)}=\rho_{a'a'}^{(0)}$, $\rho_{bb}^{(0)}=\rho_{cc}^{(0)}$, i.e., we consider the case of equal population of the upper (b and c) and lower (a and a′) working levels. Moreover, we assume that

$$\gamma_a=\gamma_{a'}=\gamma_b=\gamma_c=\Gamma,\ \gamma_{ab}=\gamma_{a'c}=\gamma.\qquad (14)$$

The equations in (11) differ from those in (10) only by the coefficient in front of V in the last equations. However, (11) can be reduced to (10) by overdetermining V (or, equivalently, the dipole moment μ). In fact, substituting in (11)

$$V=\sqrt{3/2}\hat{V},\ U=\sqrt{2/3}\hat{U},\qquad (15)$$

we reduce (10) and (11) to an identical form.

We now note that, in the E^3 approximation, the overdetermination of V and U does not affect the limits of the subdivision of the frequency scale into physically different regimes (standing and traveling-wave regimes) of generation in the ring laser. The limits of these intervals are determined only by the detuning δ and the constants of the medium. This fact is, in particular, a consequence of the material equations for the medium, taken as a mixture of the isotopes of the gases.

3. We shall suppose that the laser medium is a mixture of the isotopes of two gases. The field in the ring laser will be written in the form of two traveling waves

$$E(t,\ x)=E_1(t)\cos(\nu t+\varphi_1(t)+kx)+E_2(t)\cos(\nu t+\varphi_2(t)-kx),\qquad (16)$$

where $E_i(t)$ and $\varphi_i(t)$ are slowly-varying amplitudes and phases, and $\nu+\dot{\varphi}_i(t)$ and k are the frequencies and wave vectors of the waves.

The rate of change of the wave amplitudes E_i is then given by[3]

$$\dot{E}_i+\frac{1}{2}\frac{\nu}{Q}E_i=\frac{1}{2}\frac{\nu}{Q}R\left\{1-I_i-I_j+\frac{1}{2}[f(\omega)+f(\Omega)]I_j\right\}E_i.\qquad (17)$$

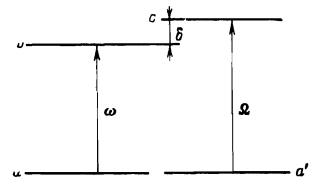

FIG. 2

Here, Q is the quality factor of the cavity, R is the intensity excess above the threshold value $R_0 = 1$, and $I_i = \mu^2E_i^2/2\hbar^2\gamma\Gamma$ are the dimensionless traveling-wave intensities. The parameter

$$f(\omega)=-\gamma\frac{\Gamma}{(ku)^2}+\frac{(\omega-\nu)^2/\gamma^2}{1+[(\omega-\nu)/\gamma]^2}\qquad (18)$$

represents the spatial (first term) and spectral (second term) hole burning of the medium.

Equation (17) describes a gas with split energy levels (Fig. 1) and the case of a gas consisting of two isotopes with partial pressure ratio of 1 : 1 (Fig. 2). When the partial amounts of the isotopes are in the ratio m : n (m + n = 1), Eq. (17) can be rewritten in the form

$$\dot{E}_i+\frac{1}{2}\frac{\nu}{Q}E_i=\frac{1}{2}\frac{\nu}{Q}RE_i\{1-I_i-I_j+[mf(\omega)+nf(\Omega)]I_j\}.\qquad (19)$$

Since the nonlinear terms in the polarization, which correspond to different line components, are additive, the expression given by (19) with $m \neq n$ corresponds to laser generation for atoms with different b → a and c → a transition probabilities. Analysis of (19) shows that when

$$mf(\omega)+nf(\Omega)<0\qquad (20)$$

laser generation occurs on a single traveling wave, whilst in the opposite case we have two-wave generation. The frequency band Δ corresponding to single-wave generation is given by the equation $mf(\omega) + nf(\Omega) = 0$. When $\gamma\Gamma/(ku)^2 \ll 1$, we have m = 1, n = 0 (the usual two-level system) and m = n (system with split energy levels)

$$\Delta(0)=2\gamma(\gamma\Gamma/(ku)^2)^{1/2},\qquad (21)$$

$$\Delta(\delta)=2(\gamma^2\Gamma\gamma/(ku)^2-\delta^2/4)^{1/2}.\qquad (22)$$

When $\delta = 0$, the intensity I of the laser radiation as a function of frequency ν exhibits peaks with widths given by (21) (see Fig. 3). When $\delta \neq 0$, this is not, in general, the case. When $\delta^2 < 4\gamma^2\Gamma\gamma/(ku)^2$, the topologic generation picture remains as before. When $\delta^2 > 4\gamma^2\Gamma\gamma/(ku)^2$, laser generation is sharply altered: the case of imaginary Δ corresponds to two-wave generation throughout the frequency range.

As an example, consider the case where the relaxation of the populations Γ is determined by the radiative lifetime, and the line width γ is determined by the sum of the radiation and collisional line widths: $\gamma = \Gamma + \gamma_{col}$. The width γ_{col} increases linearly with pressure p. Therefore, the quantity Δ given by (22) can, in general, be either real or imaginary. Figure 4 shows a plot of $\Delta(p)$. The cases of real and imaginary Δ can be recorded experimentally in an elementary fashion. The condition $\Delta = 0$ determines the separation δ between the spectral lines which are at least just resolved. According to (22),

$$\delta=2\gamma(\gamma\Gamma/(ku)^2)^{1/2}.\qquad (23)$$

As the pressure decreases, the homogeneous width γ can be made as close to the radiative width Γ as desired. Consequently, the resolution of close lines is

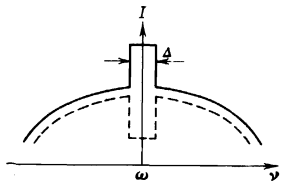

FIG. 3

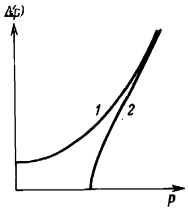

FIG. 4

FIG. 3. Generation power I as a function of frequency ν in the case of unsplit energy levels.

FIG. 4. Width of single-wave generation interval $\Delta(p)$ plotted against gas pressure p. Curve 1 corresponds to $\delta = 0$ and curve 2 to $\delta > 2\gamma[\Gamma\gamma/(ku)^2]^{1/2}$.

possible when the separation between them is smaller by a factor Γ/ku than the radiative line width. In the optical frequency band, this factor amounts to $\sim 10^{-2}$—10^{-3}. Hence, it is possible, in principle, to record the presence of fine structure in the spectral line when the separation between its components is smaller than the radiative line width by a factor of up to several hundreds.

We note that, if the line consists of more than two components, the distance between them can also be recorded provided, however, we know the number of components and their relative separation. This follows directly from a generalization of (19) and (20) to this case.

EXPERIMENTAL RESULTS

The experiment was performed with the He-Ne laser at a working wavelength of 3.39 μ. The perimeter of the three-mirror ring resonator was 120 cm. Two of the mirrors were identical, with radii of curvature of the reflecting surfaces equal to 200 cm. The mirrors were gold-coated with a reflectance of 95%. The third mirror was the exit element for the laser radiation and had a plane reflecting surface coated with a two-layer dielectric film with 97% reflectance.

In one of the arms of the resonator we placed a gas-filled tube, 35 cm long and with an internal diameter of 0.3 cm. The tube was sealed off with Brewster windows of fused quartz. The active medium was pumped in the ground state by a current, and only at high pressures of the He—Ne mixture (above 3.5 Torr) was some high-frequency power introduced to suppress discharge noise.

The two traveling waves were detected after the third mirror by two liquid-nitrogen cooled Ge—Au cells. The signals were fed into oscillographs after preliminary amplification. A typical oscillogram of the laser output for one of the traveling waves, with the resonator frequency scanned with a piezoceramic, is shown in Fig. 5. We investigated the size Δ of the region of stable single-wave operation, observed at the center of the generation line, as a function of different parameters.

The experiment showed that Δ was not very dependent on the excess of the pump intensity above the threshold value. When the radiation intensity was varied by a factor of 2, the quantity Δ increased by not more than 10%. It follows that the use of third-order perturbation theory is fully justified. The mutual scattering of oppositely traveling waves by optical surfaces inside the resonator has a definite effect on Δ [in particular, this leads to the appearance of ultra-narrow resonances at the line center, which have Lorentzian shapes and widths $\sim\gamma\Gamma\gamma/(ku)^2$]. Since the main aim of the experiment was to investigate the effect of the isotopic composition and pressure in the active medium on Δ, the resonator was tuned so that Δ was a maximum (this corresponds to minimum mode coupling through scattering), and the resonator setting was kept the same throughout each series of measurements.

Figure 6 shows the experimental dependence of Δ on the total pressure of the He—Ne^{20}—Ne^{22} mixture for three values of the partial pressure ratio m/n (0.002, 0.1, and 1). The partial pressure ratio of He to Ne was kept constant at 10 : 1. The experimental curves could be used to estimate the isotopic shift of the Ne^{20} and Ne^{22} line spectra. The form of the function $\Delta = \Delta(p)$ for the 1 : 1 ratio of Ne^{20} to Ne^{22} pressures shows that the presence of a structure in the Doppler line gives rise to the disappearance of the single-wave generation state at pressures p ~ 3.5 Torr. Substituting γ = 8(MHz) + 35(MHz/Torr), ku = 170 MHz, and Γ = 8 MHz [5] in (23), we find that the isotopic shift is $\delta \approx 50$ MHz, which is in satisfactory agreement with the results of Sakurai et al. [6]

Equations (21) and (22) enable us to investigate the presence of fine line structure by investigating the existence interval for single-wave generation. Physically, the existence of this interval is connected with the competition between spectral and spatial hole burning of the medium. The relative role of these effects is not very dependent on the parameter γ/ku and, therefore, in spite of the fact that all the above formulas were obtained on the assumption that $(\gamma/ku)^2 \ll 1$, we may expect that the qualitative conclusions with regard to the spectral width of the single-frequency generation region remain valid even when $(\gamma/ku)^2 \sim 1$.

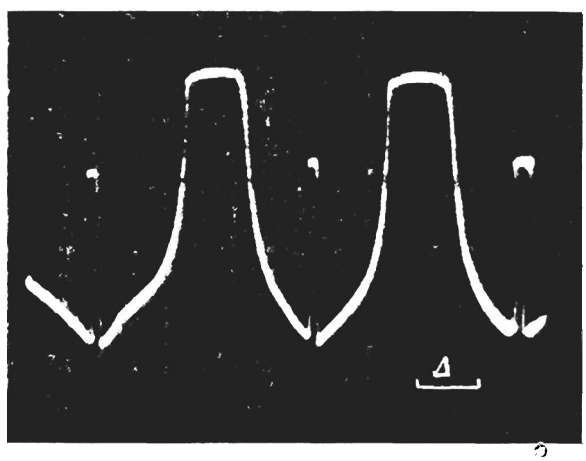

FIG. 5. Oscillogram showing the output power of one of the traveling laser waves as a function of frequency.

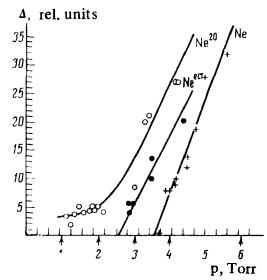

FIG. 6. Measured width of the single-wave generation interval as a function of gas pressure and isotopic composition of the mixture: ●—91 % Ne^{20} + 9% Ne^{22}, +—50% Ne^{20} + 50% Ne^{22}, ○—98% Ne^{20}.

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