Maximum resolving power of laser spectroscopy with use of frequency resonances

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It is shown that transit broadening does not decrease the resolving power of spectroscopy in which use is made of nonlinear dispersion resonances determined by varying the emission frequency of two-mode gas lasers with nonlinear absorbing cells. Using modern recording techniques, the resonance width can be 10 to 100 Hz.

PACS numbers: 06.30.Ft, 07.65. - b, 42.60.Kg

1. The broadening of reference lines in nonlinear spectroscopy, due to transit effects, is one of the main obstacles to obtaining ultranarrow resonances with widths less than a kilohertz.¹ The influence of these effects on the width and shift of power resonances of single-mode gas lasers, which fix the molecule-transition frequency in the absorbing cell, has been discussed in the literature many times.^{2–4} Recently developed experimental procedures, such as the method of separated beams,⁵ two-photon absorption,⁶ and cooling the atoms and ions in traps,⁷ promise to get rid of the harmful influence of the transit effect.

Common to all the foregoing procedures is one feature-what is registered in these experiments of absorption resonance. In the present paper we show that transit broadening does not decrease the resolving power of a spectroscopy in which nonlinear resonance dispersions are used. The resolving power is determined in this case by the collision or radiative linewidth. In Refs. 8-10, in which the nonlinear dispersion resonances were registered by measuring the change of the emission frequency of two-mode lasers with nonlinear absorbing cells, it was noted that the frequency resonances in such lasers, which are due to saturation of the dispersion of the absorbing medium, offer the advantage of high sensitivity. The fact that in this case the transit broadening does not affect adversely the resolving power of the method makes their use all the more attractive for the solution of problems in nonlinear spectroscopy and frequency stabilization.

2. The frequency of an absorbing transition in a twomode gas laser is recorded in the following manner. The difference ν of the mode frequencies of stationary gas laser operation in a stable two-mode regime is chosen such that it is known to exceed the homogeneous line width of the absorbing gas filling a cell introduced into the cavity. At the same time, this frequency difference ν is much less than the Doppler width of the line, and as a rule much less than the homogeneous gain line. The possibility of realizing stable lasing in such a regime was demonstrated in Refs. 8-10. Change of the distance between the mirrors made it possible to tune this pair of modes within the limits of the gain line.

With such a tuning, against the background of slow variation of the intermode beat frequency v, saturation of the dispersion of the absorbing gas gives rise to three narrow resonances. Two of them are produced when the frequency of one of the modes is tuned to the center of the transition line of the absorbing gas, and the third corresponds to a symmetric placement of the mode frequencies relative to the line center. For stabilization purposes, as a rule, the first two resonances are used. In this case the mode interaction in the absorbing cell is practically completely eliminated, since they interact with different groups of molecules, and the mode tuned away from the absorption line center serves as a convenient heterodyne that makes it possible to determine, from the beat frequency v, the resonant change of the frequency of the second mode located near the absorption-line center. Naturally, at the center of the resonance the second mode duplicates the frequency of the absorbing gas.

3. We assume that the field in the cavity takes the form of a standing wave with Gaussian profile

$$E(\mathbf{R}, t) = \mathscr{E}(\mathbf{R}) e^{i\omega t} + \mathbf{c.c.},$$

$$\mathscr{E}(\mathbf{R}) = \mathscr{E}_{0} \cos(kz) \exp\left[-\frac{x^{2}+y^{2}}{a^{2}}\right] = \int G(\mathbf{p}) e^{i\mathbf{p}\rho} d\mathbf{p},$$

$$G(\mathbf{p}) = \frac{a^{2}}{8\pi} \mathscr{E}_{0} \exp\left[-ikz - \frac{a^{2}p^{2}}{4}\right], \quad \mathbf{p} = (p_{x}, p_{y}),$$

$$\mathbf{p} = (x; y), \qquad k = \omega/c.$$
(1)

The influence of the absorbing cell on the frequency ω of the wave generated by the laser is described by the equation

$$\omega = \omega_L - 2\pi \omega_L \alpha', \tag{2}$$

 $\langle \mathbf{a} \rangle$

where ω_L is the mode frequency of a cavity filled with the active medium, α' is the real part of the generalized complex polarizability per unit volume of gas in the absorbing cell

$$\alpha = B^{-1} \int P(\mathbf{R}) \mathscr{E}(\mathbf{R}) d\mathbf{R}, \quad B = \int \mathscr{E}^{2}(\mathbf{R}) d\mathbf{R},$$
$$P(\mathbf{R}, t) = P(\mathbf{R}) e^{i\omega t} + \text{c.c.},$$

where P is the polarization, per unit volume, induced by the field E.

Calculation of the polarization P, accurate to terms of third order in the field inclusive, is perfectly analogous to that in Ref. 11 and leads to the following increment, quadratic in the field, to the linear polarization α_0 :

$$\alpha = \alpha_0' + i\alpha_0'' + \alpha_2' + i\alpha_2'', \tag{3}$$

$$\alpha_{2}' = -\alpha_{0}'' \frac{l}{L} \frac{\tau^{2} \varkappa^{2}}{4} \int_{0}^{\infty} d\xi \, d\eta \frac{e^{-2\xi - \eta}}{(\gamma \tau)^{2} + \xi^{2} + (\xi + \eta)^{2}} \sin\left(\frac{2\Omega}{\gamma} \xi\right),$$
(4)

$$\alpha_{2}^{"} = -\alpha_{0}^{"} \frac{l}{L} \frac{\tau^{2} \varkappa^{2}}{4} \int_{0}^{\infty} d\xi \, d\eta \frac{e^{-2\xi - \eta}}{(\gamma \tau)^{2} + \xi^{2} + (\xi + \eta)^{2}} \left[1 + \cos\left(\frac{2\Omega}{\gamma} \xi\right) \right].$$
(5)

Here *l* and *L* are the lengths of the absorbing cell and of the cavity, respectively $\tau = a/v_0$ is the time of flight of the molecule through the light beam, v_0 is the mean thermal velocity of the atom, $\kappa = |d_{12}| \mathscr{C}_0/\hbar$, where d_{12} is the matrix element of the dipole moment of the absorbing molecular transition and $\Omega = \omega - \omega_0$, where ω_0 is the frequency of the molecular transition. In Eqs. (4) and (5) it is assumed that the rate constants of the relaxation of the polarization and of the populations are the same and equal to γ , and account is taken of the fact that $|\alpha_0''| \ge |\alpha_0'|$ near the line center. In addition, in contrast to Ref. 11, the pressure was assumed to be low enough so that the relaxation of the density matrix is sufficiently accurately described by the relaxation constants, and finally, as seen from (1), no account was taken of the change in the curvature of the wave front. The absorption resonance is connected with the frequency dependence of the quantity α'' , the nonlinear increment to which, in the form (5), was first obtained in Ref. 2. The dispersion resonance is connected with the frequency dependence of the quantity $\alpha' = \alpha'_0$ $+ \alpha'_{2}$ which, as can be seen from (4), makes it also possible to fix the line center of the absorbing transition. The quantity α'_0 in (3) is a smooth function, with a width of the order of the Doppler linewidth $\Delta \omega_D$, of the frequency detuning Ω and passing through zero near the line center $\Omega = 0$. With good accuracy, therefore, in the immediate vicinity, of interest to us, of the line center, $\Omega \leq \gamma$, where the nonlinear increment to the dispersion (5) has a resonance, we can put $\alpha'_0 = 0$. It can thus be assumed that the frequency of the mode close to resonance is described by the expression $\omega = \omega_L$ $-2\pi\omega_L^{(1)}\alpha_2'$, whereas the frequency of the heterodyne mode is equal to $\alpha_h = \omega_L^{(1)} - 2\pi\omega_L^{(1)}\alpha'_0$, where $\omega_L^{(1)}$ is the frequency of the cavity filled with the active medium for the second mode. Near the resonance, the beat frequency is

$$v = \omega - \omega_h = \Delta - 2\pi \omega_L \alpha_2',$$

where

$$\Delta = \omega_L - \omega_L - 2\pi \omega_L \alpha_0',$$

and in the region $\Omega \leq \gamma$, $1/\tau$ the quantity Δ remains constant, with good accuracy, so that the resonance in the beats $\omega - \omega_h$ is determined completely by the resonant dependence of α'_2 on the detuning Ω .

4. In general outline, the dependence of α'_2 on the detuning frequency lends itself to analytic investigation. This dependence is determined by the integral

$$I' = \iint_{0}^{\infty} d\xi \, d\eta \, \frac{e^{-2\xi - \eta}}{(\gamma \tau)^2 + \xi^2 + (\xi + \eta)^2} \sin\left(\frac{2\Omega}{\gamma} \xi\right). \tag{6}$$

The integral (6) is an antisymmetric function of Ω , and we consider hereafter the case $\Omega > 0$. At $\gamma \tau \ll 1$ the denominator in (6) can be set equal to $(\gamma \tau)^2$, after which I' takes the form of the dispersion contour

$$I' = \frac{1}{2(\gamma \tau)^2} \frac{\Omega \gamma}{\gamma^2 + \Omega^2}; \quad \alpha_2' = -\alpha_0'' \frac{l}{L} \frac{\kappa^2}{8\gamma^2} \frac{\Omega \gamma}{\gamma^2 + \Omega^2}.$$

To investigate (6) in the opposite limiting case $\gamma \tau \ll 1$, following Ref. 12, we make the substitution

$$\xi = \frac{\gamma}{2\Omega} x \sin \varphi, \quad \eta = \frac{\gamma}{2\Omega} x \cos \varphi,$$

after which we obtain

$$l' = \int_{0}^{\pi/2} d\varphi \int_{0}^{\infty} \frac{\exp\left(-f\gamma x/2\Omega\right)\sin\left(x\sin\varphi\right)}{(2\Omega\tau)^2 + \psi^2 x^2} x \, dx; \tag{7}$$
$$t=2\sin\varphi + \cos\varphi, \quad \psi^2 = \sin^2\varphi + (\sin\varphi + \cos\varphi)^2.$$

At $\Omega \leq 1/\tau$, we can neglect in the denominator of the integrand of (7) the term $(\Omega \tau)^2$, after which the integral with respect to x can be calculated. Making the substitution $1 + \cot \varphi = z$, we obtain

$$I' = \int_{1}^{\infty} \frac{dz}{z^2 + 1} \operatorname{arctg}\left(\frac{2\Omega}{\gamma} \frac{1}{z + 1}\right), \quad \Omega \ll \frac{1}{\tau}.$$
 (8)

The integral in (8) at $\Omega \ll \gamma$ is a linear function of the detuning

$$I' = \frac{2\Omega}{\gamma} \cdot \frac{1}{4} \left(\frac{\pi}{2} - \ln 2 \right),$$

and reaches a constant value $I' = \pi^2/8$ at $\Omega \gg \gamma$. Thus, in the wide region $\gamma \ll \Omega \ll 1/\tau$ the integral (7) remains practically constant at $I' = \pi^2/8$.

At $|\Omega| \gtrsim 1/\tau$ we can neglect the exponential under the integral sign in (7), after which we get

$$I' = \frac{\pi}{2} \int_{1}^{\infty} \exp\left[-\frac{2\Omega\tau}{(1+z^2)^{\frac{1}{2}}}\right] \frac{dz}{1+z^2}, \quad |\Omega\tau| \ge 1.$$
(9)

In the region $|\Omega \tau| \ge 1$ we get from (9) the dispersion dependence on the detuning

$$I' = (\pi/2) (2\Omega \tau)^{-1}$$

whereas when Ω tends formally to zero, Eq. (9) yields a value $I' = \pi^2/8$, which coincides with the limiting value of (8) at $\Omega \ge \gamma$.

It is clear from the foregoing that the total dependence of (8) on the detuning frequency Ω at $\gamma \tau \ll 1$ can be obtained by interpolating Eqs. (8) and (9). In this case the integral I' in a narrow region of the frequencies $0 < \Omega \leq \gamma$ reaches its limiting value $I' = \pi^2/8$ and remains practically constant in a wide region of values $\gamma \leq \Omega < 1/\tau$, after which it decreases at $\Omega \gtrsim 1/\tau$ in accordance with (9). It is important that the rate at which (6) reaches a limiting value $I' = \pi^2/8$ does not depend on the transit time τ and is determined only by the collision line width γ . Figure 1 shows the $I'(\Omega)$ dependence obtained by numerical calculation for the values $\gamma \tau = 0.1$ and $\gamma \tau = 0.01$. The dashed curves in this figure show the values of $I'(\Omega)$ obtained from Eqs. (8) and (9).

In experiment one usually determines not the function $I'(\Omega)$ itself, but the derivative $dI/d\Omega$ with respect to the de-



FIG. 1. Shapes of frequency resonances for two values of the parameter $\gamma\tau$ (solid curves): 1) $\gamma\tau = 0.1$; 2) $\gamma\tau = 0.01$. Dashed lines—asymptotes of expression (8), which does not depend on $\gamma\tau$, and (9) for the same value of the parameter $\gamma\tau$.

tuning frequency. Figure 2 shows the dependence of $dI/d\Omega$ on Ω at $\gamma \tau = 0.01$ for the case when the line consists of two components of equal intensity, separated in frequency by $\delta = 2\gamma$. Even though the splitting δ in this case is smaller by a factor of 50 than the reciprocal transit time of the molecule through the light beam, the fine structure of the line is clearly pronounced in the figure.

Thus, transit broadening does not influence the resolving power, and the picture hardly differs from that given by two dispersion-shape components of width γ . Naturally, these valuable properties of the dispersion resonance stem from the contribution made by the slow molecules, which interact for a long time with the light beam. The contribution of these molecules is concentrated in the narrow frequency region $\Omega \leq \gamma$ and in the integral sense is small, but it turns out to be decisive for the resolving power of the method.

The role of the slow molecules manifests itself also in the sharpening of the absorption resonance, something repeatedly mentioned in the literature.^{3,4,12} In particular, such a sharpening has made it possible to observe experimentally⁴ the hyperfine structure of a line. There is, however, a qualitative difference in the possibility of experimentally using this sharpening, which we must stop to discuss. The frequency dependence (5) is determined by an integral of the form

$$I'' = \int_{0}^{\pi/2} d\varphi \int_{0}^{\infty} \frac{\exp\left(-fx\gamma/2\Omega\right) \left[1 + \cos\left(x\sin\varphi\right)\right] x \, dx}{(2\Omega\tau)^2 + \psi^2 x^2}, \qquad (10)$$

the behavior of which at $\gamma \tau \ll 1$ in the frequency region $\Omega \ll 1/\tau$ can be investigated in the following manner. We consider the integral



FIG. 2. Dependence of the first derivative of the frequency resonance on the detuning ($\gamma \tau = 0.01$) for the case when the line consists of two components separated by $\delta = 2$ (the transit broadening is 50 times larger than δ).

$$J = \int_{0}^{\infty} \frac{e^{-\alpha z - iqx}}{\beta^{2} + x^{2}} x \, dx = \ln \frac{1}{\beta} + \frac{1}{2} \int_{0}^{\infty} \ln \left[\beta^{2} + \frac{(\alpha - iq)^{2}}{(\alpha^{2} + q^{2})^{2}} z^{2} \right] e^{-z} \, dz.$$
(11)

The integral in the right-hand side of (11) converges at $\beta = 0$, therefore at $\beta < 1/\alpha$ and at any q we can put $\beta = 0$, after which we obtain

Re
$$J = \ln \frac{1}{\beta} - C - \frac{1}{2} \ln (\alpha^2 + q^2), \quad C \approx 0.577.$$

Substitution of this expression in (10) and integration with respect to φ leads to the result

$$I'' = F - \frac{1}{2}Q(\Omega),$$

$$F = \frac{\pi}{4} \ln \frac{1}{\gamma \tau} - \int_{1}^{\infty} \frac{dz}{1+z^{2}} \ln \left(\frac{1+z}{(1+z^{2})^{1/2}} e^{c} \right),$$

$$Q(\Omega) = \int_{1}^{\infty} \frac{dz}{1+z^{2}} \ln \left[1 + \frac{1}{(z+1)^{2}} \frac{4\Omega^{2}}{\gamma^{2}} \right].$$
(12)

This expression coincides with that used in Ref. 3 to estimate the half-width Γ of the resonance, which was obtained from the requirement

$$I''(\Omega=\Gamma)=1/2I''(\Omega=0)$$

and which was found to be $\Gamma \approx 1.51(\gamma/\tau)^{1/2}$. Since the function of the frequency $Q(\Omega)$ is independent of τ , the dependence of the resonance half-width on τ is due to the presence in (12) of a background *F*, independent of frequency, that increases with decreasing parameter $\gamma\tau$. If it is assumed that the experimental procedure of fixing the resonance makes it possible to eliminate this background, for example if one measures in the experiment the derivative $dI''/d\Omega = -(1/2)dQ/d\Omega$, the situation becomes perfectly analogous to that considered above for the dispersion resonance, in which case there is in principle no background. Thus, if it were possible to eliminate in (12) the background, it would be possible to state that the absorption resonance, just as that of the dispersion, is free of the transit broadening. In this connection we must note the following.

When account is taken of (12), the absorption resonance α_0'' takes at $\gamma \tau \ll 1$ the form

$$\alpha_0'' = \alpha_0 \left[1 - \frac{\tau^2 \varkappa^2}{4} \left(F - \frac{1}{2} Q(\Omega) \right) \right], \quad \Omega \tau \leq 1.$$
 (13)

The presence in the function $Q(\Omega)$ of a resonance with collision width γ is due to the contribution of the slow molecules, and in order for this resonance not to broaden as a result of saturation it is necessary that the saturating power be sufficiently low, that is, $(1/4)(dE/\hbar\gamma)^2 \leq 1$. Determining from this condition the limiting saturating power $(dE/\hbar)^2 = 4\gamma^2$ and substituting in (13), we obtain

$$\alpha_0'' = \alpha_0 \left[1 - \tau^2 \gamma^2 \left(F - \frac{1}{2} Q(\Omega) \right) \right] \approx \alpha_0 \left[1 + \frac{\tau^2 \gamma^2}{2} Q(\Omega) \right].$$

At $\tau \gamma = 0.01$ the intensity of the $Q(\Omega)$ resonance is smaller by a factor 10^4 than the intensity of the background. At such a small ratio of the intensity of the useful signal to the background one can hardly hope to record this signal in experiment.

5. The condition that there be no field-induced broadening for the slow molecules can be satisfied also in the case of dispersion resonance. Assuming $\kappa^2 = (dE/\hbar)^2 = 4\gamma^2$ and $\gamma = qA$, where $A = 4\omega^3 d^2/3\hbar c^3$ is the probability of the radiative transition and the coefficient $q \ge 1$ shows by how many times the homogeneous line width is larger than the radiative width, we obtain for the saturating power G_{sat} $= cE_{\text{sat}}^2/8\pi$

 $G_{\rm sat} = (8\pi/3)\hbar\omega A q^2/\lambda^2$,

where λ is the radiation wavelength.

We present a numerical estimate of the possibility of observing a resonance with a width less than the transit width for the case of the $F_2^{(2)}$ component of the vibrational-rotational transition P (7) of the v_3 band of methane $(\lambda = 3.38 \,\mu\text{m})$, which is of particular interest for frequency stabilization. Radiative probability of the transition is $A = 10 \,\text{sec}^{-1}$. We denote by N the gas density at which the collision line width $\Gamma(\gamma = \Gamma + A)$ becomes comparable with the radiative width. Bearing in mind that both the collision line width and the linear absorption coefficient $k = 4\pi \,(\omega/c) \,\alpha_0^{"}$ are proportional to the gas density N, we obtain

$$G_{\text{sat}} = \frac{8\pi}{3} \hbar \omega \frac{A}{\lambda^2} \left(\frac{N}{N_0}\right)^2;$$
$$(\omega - \omega_h)_{\text{max}} = \frac{ck}{2} (\tau\gamma)^2 = \frac{1}{2} ck_0 (\tau A)^2 \left(\frac{N}{N_0}\right)^3;$$

where $(\omega - \omega_h)_{\text{max}}$ is the resonance swing and k_0 is the absorption coefficient at $N = N_0$. Thus, the level of the saturating power at which the field broadening does not mask the collision broadening of the resonances decreases with density in proportion to N^2 , while the resonance itself in proportion to N^3 .

Assuming the broadening constant to be $d\Gamma/dp = 10^8$ sec⁻¹·Torr⁻¹, the absorption $dk/dN = 0.1 \text{ cm}^{-1}$ ·Torr⁻¹, and $\tau = a/v_0 = 2 \cdot 10^{-4}$ sec $(a = 10 \text{ cm}, v_0 = 5 \cdot 10^4 \text{ cm/sec})$, we find that to separate a resonance of width $\gamma = 12A$ ($\gamma/2\pi \approx 20 \text{ Hz}$) the required power level is $G_{\text{sat}}^{(1)} \sim 10^{-8} \text{ W/cm}^2$; in this case the resonance swing is $(\omega - \omega_h)_{\text{max}}^{(1)} \sim 1 \text{ Hz}$. The same values for resonances with $\gamma = 60A$ ($\gamma/2\pi = 10^2 \text{ Hz}$) are respectively $G_{\text{sat}}^{(2)} \sim 2 \cdot 10^{-7} \text{ W/cm}^2$ and $(\omega - \omega_h)_{\text{max}}^{(2)} \sim 10^2 \text{ Hz}$. It can be seen that the condition for the level of the saturating power is quite stringent and its realization calls for special devices that lower the power density inside the absorbing cell. We emphasize, however, that this condition is not peculiar to the considered recording method; the problem of field broadening in recording such narrow resonances is encountered in the same form in any procedure. As for the two-mode method, experiments performed in Refs. 8 and 9 show that such a low field density in the absorbing cell is realistic. Indeed, the saturating power in the absorbing cell in Refs. 8 and 9, at a = 1.5 cm and using a special procedure for lowering the intensity of one of the modes, was $G_{sat}^{(0)} \sim 10^{-6}$ W/cm², i.e., to obtain the required level $G_{sat} \sim 10^{-8}$ W/cm² it suffices to increase the transverse dimension of the field by 10 times ($a \approx 15$ cm), which is realistic (fields of such sizes were already realized for single mode lasers^{13,14}).

The maximum sensitivity of separating resonances in the considered two-mode method is determined by the level of the natural frequency fluctuations of the laser radiation,⁸⁻⁹, i.e., it amounts to $\sim 10^{-1}$ Hz/Hz^{1/2}. This means that to record resonances of width $\gamma/2\pi = 20$ Hz at a signal/ noise ratio $S/N = 10^2$ the integration constant of the signal should be $t \int = 20$ sec. As for the resonances of width $\gamma/2\pi = 10^2$ Hz, from the presented estimate of $(\omega - \omega_h)_{\text{max}}^{(2)}$ one can see that they can be reliably recorded $(S/N = 10^2, t \int = 0.2 \text{ sec})$ even at a sensitivity ~ 1 Hz/Hz^{1/2}, which was experimentally realized in Refs. 8 and 9, and which is approximately lower by one order than the maximum value.

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Translated by J. G. Adashko