

Theory of one-photon high-resolution absorption optical spectroscopy*

J. I. Cotrim Vasconcellos[†]

Department of Physics and Electrical Engineering, University of Southern California, Los Angeles, California 90007
Instituto de Física "Gleb Wataghin," Universidade Estadual de Campinas, C. P. 1170, Campinas, S. P. Brasil

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We consider in this paper the Doppler-broadened absorption by two-level molecules of a gas from a weak traveling electromagnetic plane wave in the presence of an oppositely traveling strong plane wave at the same frequency. Here the absorption spectrum exhibits a dip that broadens and deepens as the gas is increasingly saturated by the strong wave. This saturation effect has been employed in the recent experiments on the fine structure of atomic hydrogen performed by Hänsch, Shahin, and Schawlow (HSS). The form, linewidth, and strength of saturation dips in the fine-structure lines are studied for experimental parameters appropriate to the hydrogen experiment. In addition, saturation line-shape curves are calculated for experimental situations in which the strengths of the stronger field give rise to stronger or weaker saturation of the gas medium and varying degrees of Doppler broadening exist. The calculation of shape of the saturation dip and absorption curves for a Doppler-broadened gas of two-level atoms is made exactly; in effect, the strong field is treated as arbitrarily strong by avoiding a perturbation-theory treatment in favor of the exact solutions for a driven two-level system. The general behavior found is in accord with intuition and experimental observations by HSS. There is an optimum saturation power which gives a prominent dip and beyond which the dip suffers "power broadening."

I. INTRODUCTION

In absorption experiments in gas systems within the visible and infrared spectral ranges, the broadening of the absorption lines is predominantly Gaussian for low gas pressures and becomes a mixture of Lorentzian and Doppler broadenings as the pressure increases.

In the new field of optical saturation, or "Lamb dip," spectroscopy, one is able to study atomic and molecular fine and hyperfine spectral components because Doppler broadening is avoided. The normal Doppler-broadened line shapes have a width of the order of 10^8 or 10^9 Hz, at room temperatures, and so spectral details normally become obscured by the Doppler broadening.

The possibility of using saturation of a resonance in various ways to increase resolution of inhomogeneously broadened lines was suggested early by Portis¹ for nuclear and paramagnetic resonance and by Javan² for optical spectroscopy. Many variations of saturation techniques are possible, such as those of Brewer, in which the line is moved while the exciting frequency is fixed,^{3,4} rather than scanning a fixed transition with a tunable frequency, the case we consider in this paper.

The idea is to "burn a hole" in the inhomogeneously broadened spectral lines. This is achieved by making the molecules of the gas interact with a standing wave inside the laser cavity or interact with two opposite traveling waves. The molecules that will interact on-resonance with both beams simultaneously are just the ones with zero-velocity

components along the direction of propagation of the waves, i.e., just those molecules that do not have a Doppler shift in their resonance angular frequency ω_0 .

With this technique, a great deal of information about atomic and molecular structure has been obtained. In a relatively short period of time (since 1969) several important results, such as resolving the fine structure of atomic hydrogen⁵ and resolving the hyperfine structure of sodium vapor,⁶ were obtained. Also, it opened the possibility of studying new coherent spectroscopy effects, with a number of applications.⁴ Two excellent reviews of these achievements are those by Brewer⁴ and by Shimoda and Shimizu.³

We consider in this work the nonlinear absorption of a weak electromagnetic test wave by two-level molecules in the presence of a strong electromagnetic wave at the same frequency, but traveling opposite to the weak wave. We calculate the power density absorbed from the weak wave, predicting from first principles the absorption for several saturation conditions (Sec. II). The relaxation mechanisms are radiation damping and hard collisions. Since in the experiment done by Hänsch, Shahin, and Schawlow⁵ the pressure is taken as low as 0.1–1 Torr, and in general pressure effects are avoided in high-resolution experiments, we assume that the Doppler broadening predominates over the homogeneous broadening. The thermal motion of the molecules of the gas is taken into account, in the case in which the Doppler broadening predominates over the homogeneous broadening.

II. POWER DENSITY ABSORBED FROM THE OPTICAL WEAK WAVE BY A GAS MEDIUM OF TWO-LEVEL MOLECULES IN THE PRESENCE OF A STRONG OPTICAL FIELD, WITH THE SAME FREQUENCY

A. Basic assumptions: The theoretical model for the gas

(1) The gaseous medium is composed of molecules that are supposed initially to interact weakly with each other. Hence each molecule can be represented by a statistical ensemble of identical molecules in the thermal bath composed by the others. The condition of near-resonance with the external fields allows us to consider the molecule as a two-level system. We also assume that the two levels of interest are sufficiently separated from other energy levels, so that the two-level approximation is valid.

The external optical electric fields to be considered are supposed to be monochromatic oscillating plane waves,

$$\vec{E}(\vec{r}, t) = \text{Re}[\vec{E}_0(\vec{r})e^{i(\omega_L t - \vec{k} \cdot \vec{r})}], \quad (2.1)$$

where ω_L is the angular frequency of the incoming wave and \vec{k} is the wave vector.

We assume, as usual, that this field varies in space very slowly over the dimensions of the molecule, and we restrict our treatment to the linear dipole approximation for the light-gas interaction. Then, the interaction Hamiltonian is written

$$V = -\vec{\mu} \cdot \vec{E}, \quad (2.2)$$

where $\vec{\mu}$ is the electric-dipole-moment vector of the molecule (or atom) and \vec{E} is given by (2.1).

(2) As is well known, several factors contribute to the finite linewidth of spectral lines of gases. Some of them are related to relaxation processes of the energy levels of the molecule or atom causing homogeneous broadening. Of those we consider here spontaneous emission and collisions between the molecules. Other factors to be considered here are saturation broadening due to the high strengths of the applied external fields, and the Doppler broadening caused by the thermal motion of the molecules. The saturation mechanism gives a homogeneous contribution to the absorption linewidths and the Doppler effect an inhomogeneous one.

The collisions are studied in the so-called impact approximation, since we are dealing with low-pressure regimes. We take into account not just the pressure broadening caused by the collisions of the molecules with each other, but also the collisions with the walls of the gas container.⁷ In the impact approximation, the collisions are assumed to occur in a random way and to be so strong that immediately after they take place all phase information is lost ("hard" or strong collisions).

This character of randomness has its counterpart in spontaneous emission. If a quantum system is in one of its excited energy states, the radiation decay occurs at random and the probability of spontaneous emission obeys a Poisson distribution.

The interaction of the molecule with the external fields can be interrupted either by a strong collision or by a spontaneous emission in the present context. This assumption, plus the fact that radiation damping and hard collisions are statistically independent, allow the formulation of an unified approach to them.

Let $f(\theta)$ be the probability that a molecule will not be interrupted during the time interval θ . It is well known that

$$f(\theta) = e^{-\theta/\tau}, \quad (2.3)$$

where τ is the homogeneous relaxation time, i.e., the average time between two successive interruptions. We write

$$\frac{1}{\tau} = \frac{1}{\tau_c} + \frac{1}{\tau_n}, \quad (2.4)$$

where τ_c is the average time between two successive collisions and τ_n is the average time between two successive spontaneous decays.

The probability that a molecule, after surviving without interruptions for a time θ , suffers an interruption in the time interval between θ and $\theta + d\theta$ is

$$g(\theta) d\theta = e^{-\theta/\tau} d\theta/\tau, \quad (2.5)$$

which has the form of a Poisson distribution.

B. Calculation of the absorbed power density

In 1957, Feynman, Vernon, and Hellwarth⁸ derived a geometrical representation for the Schrödinger equation of an ensemble of two-quantum-level noninteracting systems which are under the influence of a perturbation. We use this vector model for calculating the power density absorbed from the weak wave by a gas medium of two-level molecules in the presence of a strong field. We then calculate the variation of internal energy of the atomic (or molecular) two-level system due to its absorption of light from the weak wave. According to the vector model, the absorbed power density from the weak wave is

$$\frac{dW}{dt} = N \frac{\hbar\omega_0}{2} \frac{d}{dt}(\delta r_3), \quad (2.6)$$

where N is the particle density, r_3 is the third component of the vector model's vector \vec{r} in the presence of the strong field only, and δr_3 is the modification of this component due to the presence of the weak field.

The two optical waves applied to the gas medium

have an electric field given by

$$E = E_1 \cos(\omega_- t - \phi_0) + E_2 \cos(\omega_+ t + \phi_0), \quad (2.7)$$

where $\omega_- = \omega_L(1 - v/c)$ and $\omega_+ = \omega_L(1 + v/c)$ are the Doppler-shifted frequencies of the waves seen from the frame of reference of the molecule, ω_L is the laser frequency in the laboratory frame, $\phi_0 = kz_0$ is a phase, and E_1 and E_2 are the amplitudes of the two waves. It is assumed that $E_1 \gg E_2$. From its own frame of reference, a molecule "sees" two waves of frequencies ω_- and ω_+ (molecule's system of reference M).

Suppose that in the presence of the strong field alone the vector representing the state of the two-level system is \vec{r} , and $\delta\vec{r}$ is its modification in the presence of the weak wave. The total vector of the two-level system is

$$\vec{R} = \vec{r} + \delta\vec{r}.$$

Similarly, the vector $\vec{\Omega}$ representing the perturbation is written

$$\vec{\Omega} = \vec{\omega} + \delta\vec{\omega}.$$

The equation of motion for \vec{R} is

$$\frac{d\vec{R}}{dt} = \vec{\Omega} \times \vec{R}. \quad (2.8)$$

First, we obtain the solution for \vec{r} , i.e., the solution in the absence of the weak wave. The equation to be solved is

$$\frac{d\vec{r}}{dt} = \vec{\omega} \times \vec{r}, \quad (2.9)$$

where the vector $\vec{\omega} = (\omega_1, \omega_2, \omega_3)$ is given by its components,

$$\begin{aligned} \omega_1 &= (1/\hbar)(V_{12} + V_{21}) \\ &= -(2\mu_{12}/\hbar)E_0 \cos(\omega_- t - \phi_0), \end{aligned} \quad (2.10)$$

$$\omega_2 = (i/\hbar)(V_{12} - V_{21}) = 0,$$

$$\omega_3 = \omega_0,$$

for the perturbation matrix elements given by

$$V_{12} = V_{21} = -\mu_{12}E_0 \cos(\omega_- t - \phi_0),$$

$$V_{11} = V_{22} = 0.$$

To simplify the calculations, we change to a rotating coordinate system M' that rotates with frequency ω_- around the axis 3 in the \vec{r} space. The rotation vector is given then by $\vec{\omega}_- = (0, 0, \omega_-)$. The rotation matrix R is

$$R = \begin{bmatrix} \cos\omega_- t & \sin\omega_- t & 0 \\ -\sin\omega_- t & \cos\omega_- t & 0 \\ 0 & 0 & 1 \end{bmatrix}. \quad (2.11)$$

The vector $\vec{\omega}$ transforms as $\omega'_i = \sum_j R_{ij}\omega_j$. This

gives, in the rotating-wave approximation,

$$\begin{aligned} \omega'_1 &= -(\mu_{12}E_0/\hbar) \cos\phi_0, \\ \omega'_2 &= +(\mu_{12}E_0/\hbar) \sin\phi_0, \\ \omega'_3 &= \omega_0. \end{aligned} \quad (2.12)$$

Similarly, we have

$$R \begin{bmatrix} r_1 \\ r_2 \\ r_3 \end{bmatrix} = \begin{bmatrix} r'_1 \\ r'_2 \\ r'_3 \end{bmatrix} = \begin{bmatrix} \text{Re}[(r_1 + ir_2)e^{-i\omega_- t}] \\ \text{Im}[(r_1 + ir_2)e^{-i\omega_- t}] \\ r_3 \end{bmatrix}. \quad (2.13)$$

In a frame rotating with angular velocity ω_- , $d\vec{r}/dt$ transforms into $d\vec{r}'/dt + \vec{\omega}_- \times \vec{r}'$. Hence the equation of motion for \vec{r}' in the M' system of reference becomes

$$\frac{dr'_1}{dt} = (\omega_- - \omega_0)r'_2 + \omega'_2 r'_3,$$

$$\frac{dr'_2}{dt} = -(\omega_- - \omega_0)r'_1 - r'_3 \omega'_1,$$

$$\frac{dr'_3}{dt} = \omega'_1 r'_2 - \omega'_2 r'_1.$$

For solving this system of equations, we use the Laplace transform method, using as initial conditions

$$r'_1(\Theta = 0) = r'_2(\Theta = 0) = 0, \quad r'_3(\Theta = 0) = -1,$$

where $\Theta = t - t'$, t' being the time of last interruption by collision or spontaneous decay. This initial condition means that the atom (or molecule) is in the lower state of energy in the two-level system. To perform the Laplace transform we change the independent variable t to $\Theta = t - t'$, where t' is the time of last collision, and get

$$\begin{aligned} r'_1(S) &= (1/\Delta)[(\omega_- - \omega_0)\omega'_1 - \omega'_2 S], \\ r'_2(S) &= (1/\Delta)[\omega'_2(\omega_- - \omega_0) + S\omega'_1], \\ r'_3(S) &= -(1/\Delta)[S^2 + (\omega_- - \omega_0)^2], \end{aligned} \quad (2.14)$$

where S is the Laplace transform parameter and Δ is given by

$$\begin{aligned} \Delta &= \begin{bmatrix} S & -(\omega_- - \omega_0) & -\omega'_2 \\ (\omega_- - \omega_0) & S & \omega'_1 \\ \omega'_2 & -\omega'_1 & S \end{bmatrix} \\ &= S(S^2 + \Omega^2), \end{aligned}$$

with

$$\Omega^2 = (\omega_- - \omega_0)^2 + \mu_{12}^2 E_0^2 / \hbar^2.$$

Inverting the Laplace-transformed components of \vec{r} , we find

$$\begin{aligned} r'_1(\Theta) &= \frac{\omega'_1(\omega_- - \omega_0)(1 - \cos\Omega\Theta)}{\Omega^2} - \frac{\omega'_2}{\Omega} \sin\Omega\Theta, \\ r'_2(\Theta) &= \frac{\omega'_2(\omega_- - \omega_0)(1 - \cos\Omega\Theta)}{\Omega^2} + \frac{\omega'_1}{\Omega} \sin\Omega\Theta, \\ r'_3(\Theta) &= -1 + (\mu_{12}^2 E_0^2 / \hbar^2 \Omega^2)(1 - \cos\Omega\Theta). \end{aligned} \quad (2.15)$$

Next we obtain the solution for $\delta\vec{r}$ in the approximation in which the term $\delta\vec{\omega} \times \delta\vec{r}$ in (2.9) is neglected. Then, using

$$\frac{d}{dt}(\delta\vec{r}) = \vec{\omega} \times \delta\vec{r} + \delta\vec{\omega} \times \vec{r}, \quad (2.16)$$

with the solutions obtained for the δr_1 and δr_2 components of the vector $\delta\vec{r}$, we obtain the part of $d(\delta r_3)/dt$ that is quadratic in the strength of the weak field E_2 , namely,

$$\frac{d}{dt}(\delta r_3) = \delta\omega_1 \delta r_2 - \delta\omega_2 \delta r_1. \quad (2.17)$$

The equations of motion for the components of $\delta\vec{r}$ in the rotating frame M' are

$$\begin{aligned} \frac{d}{dt}(\delta r'_1) &= \omega'_2 \delta r'_3 + (\omega_- - \omega_0) \delta r'_2 + \delta\omega'_2 r'_3, \\ \frac{d}{dt}(\delta r'_2) &= -(\omega_- - \omega_0) \delta r'_1 - \omega'_1 \delta r'_3 - \delta\omega'_1 r'_3, \\ \frac{d}{dt}(\delta r'_3) &= \omega'_1 \delta r'_2 - \omega'_2 \delta r'_1 + \delta\omega'_1 r'_2 - r'_1 \delta\omega'_2, \end{aligned} \quad (2.18)$$

where the prime indicates the rotated quantities, Eq. (2.13), and

$$\begin{aligned} \omega'_1 &= -\frac{\mu_{12} E_1}{\hbar} \cos\phi_0, & \omega'_2 &= +\frac{\mu_{12} E_1}{\hbar} \sin\phi_0, \\ \delta\omega'_1 &= -(\mu_{12} E_2 / \hbar) \cos[(\omega_+ - \omega_-)t + \phi_0], \\ \delta\omega'_2 &= -(\mu_{12} E_2 / \hbar) \sin[(\omega_+ - \omega_-)t + \phi_0], \end{aligned} \quad (2.19)$$

in the rotating-wave approximation; ω_- and ω_+ are Doppler frequencies defined by (2.7).

The system of equations (2.18) is solved using Laplace transforms (see the Appendix). Then, the power density absorbed [Eq. (2.6)] is averaged over the homogeneous broadening mechanisms using the Poisson distribution (2.5) as the weight function:

$$\left(\frac{dW}{dt}\right)_H = \int_0^\infty e^{-\theta/\tau} \frac{d\theta dW}{\tau dt}. \quad (2.20)$$

To perform the Laplace transform we change the independent variable t to $\theta = t - t'$, where t' is the time of the last interruption (by collision or spontaneous decay). We assume the following initial conditions:

$$\delta r'_1(\theta=0) = 0, \quad \delta r'_2(\theta=0) = 0, \quad \delta r'_3(\theta=0) = 0. \quad (2.21)$$

We note, however, that because of the formal analogy between the averaging procedure used in (2.20) and the Laplace transform, it is not necessary to find the components $\delta r'_i(\theta)$ of the vector $\delta\vec{r}$ by an inverse Laplace transformation and then take the average over the homogeneous broadening processes. It is enough to choose as the Laplace transform parameter

$$S_- = 1/\tau - i\alpha, \quad (2.22)$$

where τ is the homogeneous relaxation time and $\alpha = \omega_+ - \omega_-$. This procedure represents a considerable simplification of the calculations.

After a rather lengthy calculation, where the average over the phases is also taken, we find

$$\begin{aligned} \left(\frac{dW}{dt}\right)_H &= N \frac{\hbar\omega_0}{2} \frac{\mu_{12}^2 E_2^2}{\hbar^2} \\ &\times \tau \{ \eta(s, y^2, z) [1 + (x - \frac{1}{2}z)^2] - s^2 Q \} \\ &\times \frac{1}{(1 + y^2)(1 + z^2)[(1 + y^2 - z^2)^2 + 4z^2]}, \end{aligned} \quad (2.23)$$

where

$$\begin{aligned} x &= \tau(\omega_L - \omega_0), & z &= 2(\omega_L/c)v\tau, \\ s &= (\mu_{12} E_1 / \hbar)\tau, & y^2 &= (x - \frac{1}{2}z)^2 + s^2, \\ \eta(s, y^2, z) &= (1 + y^2 - 3z^2)(1 + zx - 1.5z^2 + \frac{1}{2}s^2) \\ &\quad + z(z^2 - y^2 - 3)(x - 2.5z), \\ Q &= \frac{1}{4}(Q_1 + Q_2), \end{aligned}$$

with

$$\begin{aligned} Q_1 &= (2x - z) \{ (1 - z^2 + y^2)(x - \frac{1}{2}z) + 2z[1 - z(x - 1.5z)] \}, \\ Q_2 &= 2 \{ (1 - z^2 + y^2)[1 - z(x - 1.5z)] - 2z(x - \frac{1}{2}z) \}. \end{aligned}$$

The parameter $s = (\mu_{12} E_1 / \hbar)\tau$ is called the saturation parameter. The parameter $z = 2(\omega_L/c)v\tau$ is related to the component of the velocity of the molecule along the direction of the lasers beams. (c is the speed of light in vacuum.)

In the limit of no saturation ($s=0$), and for molecules with zero velocity v along the beam axis, we get

$$\left(\frac{dW}{dt}\right)_H = N \left(\frac{\hbar\omega_0}{2}\right) \frac{\mu_{12}^2 E_2^2}{\hbar^2} \frac{\tau}{1 + x^2}, \quad (2.24)$$

i.e., a Lorentzian curve with full linewidth at half-maximum intensity $2/\tau$, as expected.

In Figs. 1–3 the homogeneous power density $(dW/dt)_{H0}$ absorbed from the weak wave (2.23),

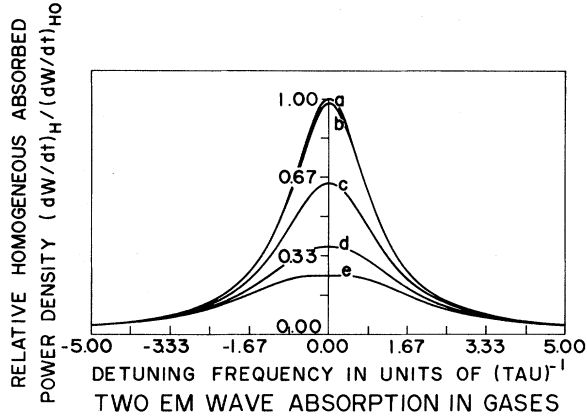


FIG. 1. Relative homogeneous absorbed power density $(dW/dt)_H / (dW/dt)_{H0}$ from the weak em wave vs detuning frequency $x = \tau(\omega_L - \omega_0)$. Absorption dependence on saturation for molecules with zero-velocity ($v = 0$) component along the laser beam; $(dW/dt)_{H0} = (dW/dt)_H$ ($s = 0$, $v = 0$, $x = 0$) $= \frac{1}{2} N \hbar \omega_0 (\mu_{12}^2 E_1^2 / \hbar) \tau$ erg/sec cm^3 ; τ is the homogeneous relaxation time (sec); saturation parameter $s = (\mu_{12} E_1 / \hbar) \tau$, assuming the values $a = 0$, $b = 0.10$, $c = 0.50$, $d = 0.80$, and $e = 1.0$.

normalized to (2.24) for $x = 0$, is shown for several values of the parameters s and z .

The final step of the calculation is to take the average over the velocities of the molecules, namely,

$$\left(\frac{dW}{dt}\right)_{\text{inhom}} = \int_{-\infty}^{+\infty} \left(\frac{dW}{dt}\right)_H f_G(\delta) d\delta, \quad (2.25)$$

where

$$f_G(\delta) = \frac{\tau_G / \tau}{\sqrt{\pi}} e^{-(\tau_G / \tau)^2 \delta^2} \quad (2.26)$$

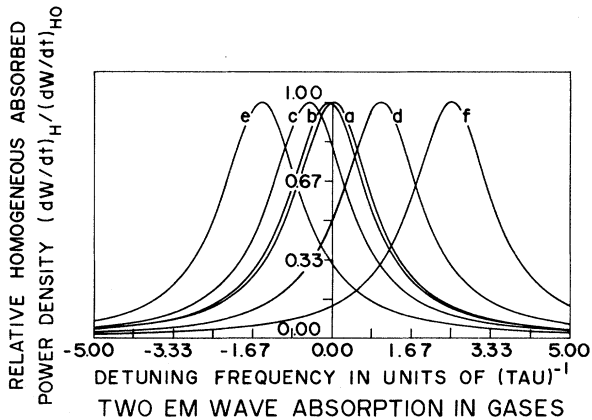


FIG. 2. Same as Fig. 1, but with absorption dependence on the velocity components v of the molecules along the laser beam, no saturation effects ($s = 0$), and $a = 0$, $b = 0.20$, $c = 1.0$, $d = -2.0$, $e = 3.0$, and $f = -5.0$ (in units of $c/2\omega_L \tau$).

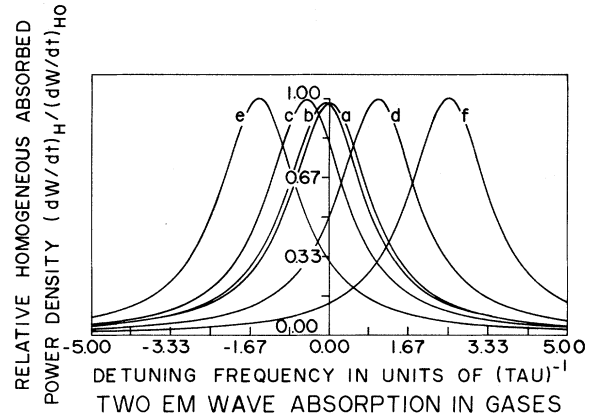


FIG. 3. Same as Fig. 2, but with saturation parameter $s = 0.10$.

is the Gaussian distribution of velocities in the units of τ , the homogeneous relaxation time.

We are interested in describing the kind of experiments of which that of Hänsch *et al.*⁵ is an example. We assume then that the linewidth of the Gaussian line, $\Delta\omega_G$, is much bigger than the homogeneous linewidth $\Delta\omega_H$. Then, the expression (2.25) can be written

$$\left(\frac{dW}{dt}\right)_{\text{inhom}} \approx f_G(\omega_L - \omega_0) \int_{-\infty}^{+\infty} \left(\frac{dW}{dt}\right)_H d\delta. \quad (2.27)$$

The integral

$$J = \int_{-\infty}^{+\infty} \left(\frac{dW}{dt}\right)_H d\delta \quad (2.28)$$

is calculated for several ratios of the homogeneous and inhomogeneous linewidths $\Delta\omega_G / \Delta\omega_L$ and for several values of the saturation parameter $s = (\mu_{12} E_1 / \hbar) \tau$. The case in which $\Delta\omega_G / \Delta\omega_H = 20$ and $s = 0.10$ represents the experimental situation of the narrowest homogeneous linewidth measured by Hänsch *et al.*,⁵ in which $\Delta\omega_H = 300$ MHz and the saturation parameter is kept small to avoid power broadening of the fine-structure line. In Figs. 4 and 5 the integral J [Eq. (2.28)] is shown for several values of the saturation parameter. In Fig. 6 we display J for $s = 0.10$ in more detail. The linewidth of the "dip" in the absorption curve obeys the expression for a saturated Lorentzian linewidth,

$$\Delta\omega_{\text{dip}} = (2/\tau)(1 + s^2)^{1/2}. \quad (2.29)$$

We could also obtain the relative depths D , of the dip for several values of the saturation parameter s . In particular, for $s = 1$ the absorption attenuates by 25% for its unsaturated value ($s = 0$).

In Fig. 7 the Doppler-broadened absorbed power density (2.27) is displayed for various values of the

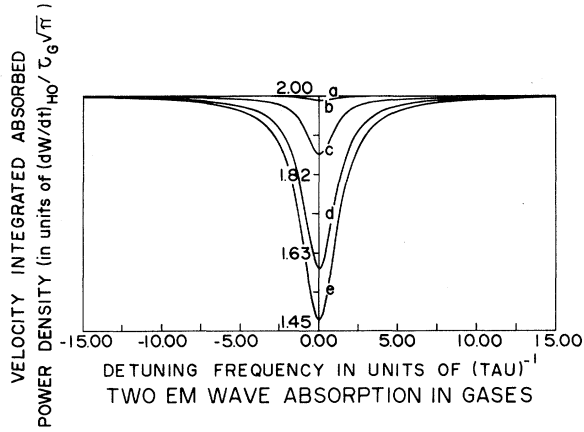


FIG. 4. Velocity-integrated absorbed power density from the weak em wave vs detuning frequency $x = \tau(\omega_L - \omega_0)$, with absorption dependence on saturation and values for the saturation parameter $s = (\mu_{12} E_1 / \hbar) \tau$ of $a = 0$, $b = 0.10$, $c = 0.40$, $d = 0.80$, and $e = 1.0$; τ_G is the Gaussian time (sec); $(dW/dt)_{H0} = (N \mu_{12}^2 E_1^2 / \hbar^2) \tau$ erg/sec cm³.

saturation parameter s , for the case in which $\Delta\omega_G / \Delta\omega_H = 20$. Figure 8 displays the theoretical results for the narrowest fine-structure component of the H_α line resolved by Hänsch *et al.*⁵ The plots in Fig. 9 represent the predicted high-resolution absorption spectra described by (2.27) for various degrees of Doppler broadening, and for the saturation parameter $s = 0.80$. In principle, these calculations can describe the high-resolution absorption spectra for any value of the satura-

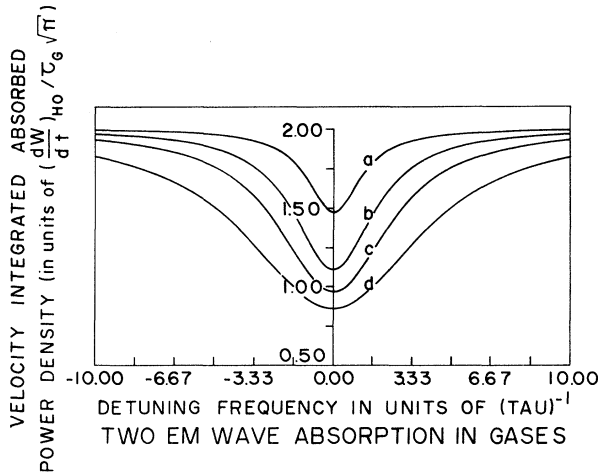


FIG. 5. Same as Fig. 4, but with absorption dependence on saturation with zero-velocity component along the laser beam, $(dW/dt)_{H0} = (dW/dt)_H$ ($s = 0$, $v = 0$, $x = 0$) $= \frac{1}{2} N \hbar \omega_0 (\mu_{12}^2 E_1^2 / \hbar^2) \tau$ erg/sec cm³ and the saturation parameter s assuming the values $a = 1$, $b = 2$, $c = 3$, and $d = 5$.

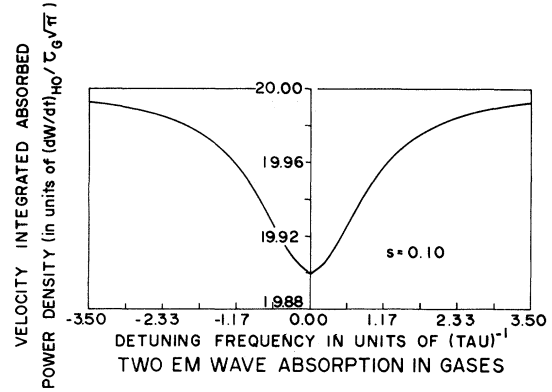


FIG. 6. Same as Fig. 5, in more detail for $s = 0.10$.

tion parameter and for experimental situations in which the inhomogeneous linewidth $\Delta\omega_G$ is much bigger than the homogeneous linewidth $\Delta\omega_H$ ($\Delta\omega_G \gg \Delta\omega_H$).

III. CONCLUSIONS

In the absorption high-resolution spectra calculated in this work there are three regions to be described. When the detuning frequency x is about zero, the homogeneous line appears without appreciable interference with the Gaussian background, since the Gaussian function is near unity. The saturation dips are Lorentzian in form, and their full linewidth at half-maximum intensity,

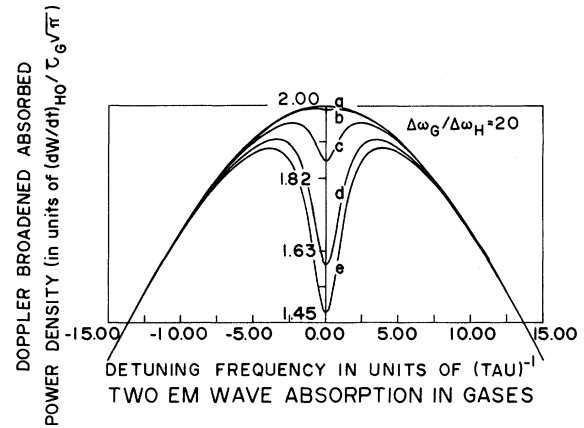


FIG. 7. Doppler-broadened absorbed power density from the weak em wave for $\Delta\omega_G / \Delta\omega_H = 20$ vs detuning frequency $x = \tau(\omega_L - \omega_0)$; $\Delta\omega_G$ is the Gaussian linewidth (in $2(\ln 2)^{1/2} / \tau_G$ units), $\Delta\omega_H$ is the Lorentzian linewidth (in $2/\tau$ units), τ is the homogeneous relaxation time (sec), τ_G is the Gaussian time (sec), $(dW/dt)_{H0} = (N \mu_{12}^2 E_1^2 / \hbar^2) \tau$ erg/sec cm³, saturation parameter $s = (\mu_{12} E_1 / \hbar) \tau$, assuming the values $a = 0$, $b = 0.10$, $c = 0.40$, $d = 0.80$, and $e = 1.0$.

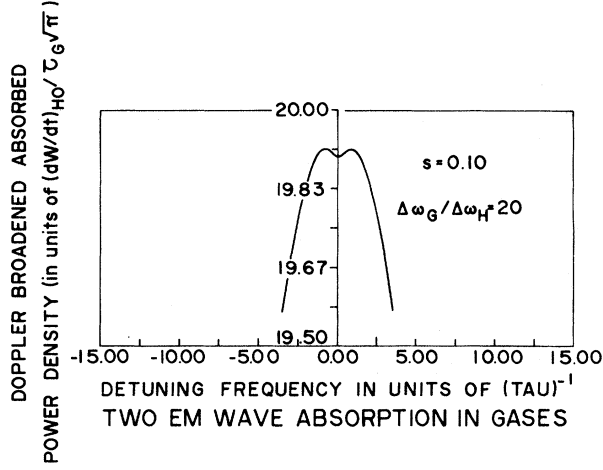


FIG. 8. Same as Fig. 7, but with the saturation parameter $s=0.10$.

$\Delta\omega_{dip}$, are given by Eq. (2.29). The strength of the dips are also predicted from the spectra calculated. We also conclude that there is an optimum saturation power which gives a prominent dip and beyond which the dip suffers "power broadening." The influence of this effect in the linewidth of the dips is described by expression (2.29).

There is an intermediate region in which the homogeneous and inhomogeneous broadening mechanism superpose and in the wing, the absorption line is purely Gaussian.

It is our hope that these calculations may lay the groundwork for further refinements which might deal, for example, with the small but important shifts in the line center known to occur in saturation spectroscopy.

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APPENDIX: SOLUTION OF EQ. (2.16) AND ITS AVERAGE OVER THE RELAXATION BROADENING MECHANISMS (HARD COLLISIONS PLUS SPONTANEOUS DECAY)

The procedure is as follows: Equation (2.16) is solved using the Laplace-transform method and a rotation of the coordinate axes. The rotation vector is $\omega_- = (0, 0, \omega_-)$. The equations of motion for the components of $\delta\vec{r}$ in the rotating frame M' are

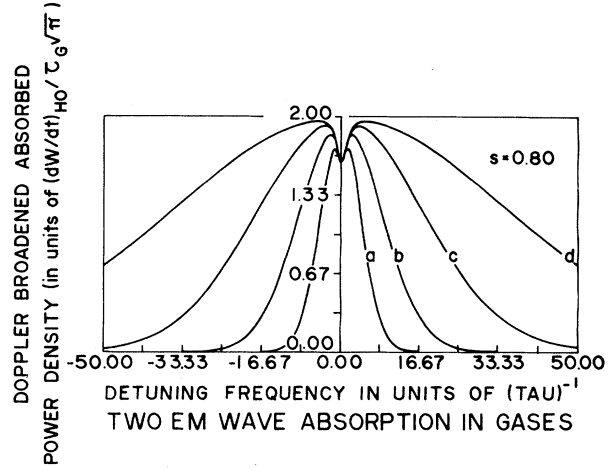


FIG. 9. Doppler-broadened absorbed power density from the weak em wave for $s=0.80$ vs detuning frequency x , with $\Delta\omega_G/\Delta\omega_H$ assuming the values $a=5$, $b=10$, $c=20$, and $d=40$; other parameters the same as in Fig. 7.

given by Eq. (2.18). The initial conditions are given by (2.21).

Taking the Laplace transform and using the initial conditions defined above, we get

$$\begin{aligned} S\delta r'_1(S) - (\omega_- - \omega_0)\delta r'_2(S) - \omega'_2\delta r'_3(S) &= a_1, \\ (\omega_- - \omega_0)\delta r'_1(S) + S\delta r'_2(S) + \omega'_1\delta r'_3(S) &= a_2, \\ \omega'_2\delta r'_1(S) - \omega'_1\delta r'_2(S) + S\delta r'_3(S) &= a_3. \end{aligned} \quad (A1)$$

S is the Laplace transform parameter:

$$\begin{aligned} \delta r'_i(S) &= \mathcal{L}[\delta r'_i(\Theta)], \quad a_1 = \mathcal{L}[\delta\omega'_2 r'_3], \\ a_2 &= \mathcal{L}[-\delta\omega'_1 r'_3], \quad a_3 = \mathcal{L}[\delta\omega'_1 r'_2 - \delta\omega'_2 r'_1], \end{aligned}$$

where \mathcal{L} is the Laplace transform.

The solution of the system of equations (A1) is given by

$$\delta r'_1(S) = \frac{1}{\Delta} \begin{bmatrix} a_1 & -(\omega_- - \omega_0) & -\omega'_2 \\ a_2 & S & \omega'_1 \\ a_3 & -\omega'_1 & S \end{bmatrix}, \quad (A2)$$

$$\delta r'_2(S) = \frac{1}{\Delta} \begin{bmatrix} S & a_1 & -\omega'_2 \\ (\omega_- - \omega_0) & a_2 & \omega'_1 \\ \omega'_2 & a_3 & S \end{bmatrix},$$

where

$$\Delta = S(S^2 + \Omega^2), \quad \Omega^2 = (\omega_- - \omega_0)^2 + \mu_{12}^2 E_1^2 / \hbar^2. \quad (A3)$$

The inverse transformation and the average over the relaxation mechanisms required in (2.20) can

be accomplished using the transform parameter (2.22). The main steps which lead to (2.23) are as follows:

$$\begin{aligned} \left(\frac{dW}{dt}\right)_H &= \int_0^\infty e^{-\Theta/\tau} \frac{d\Theta}{\tau} \left(\frac{dW}{dt}\right) \\ &= -N \frac{\hbar \omega_0}{2} \frac{\mu_{12} E_2}{\hbar \tau} (\Gamma_1 - \Gamma_2), \end{aligned}$$

where

$$\langle Z_1 \rangle_{\phi_0, \phi_1} = \frac{1}{\Delta_-} \left(S_-^2 + \frac{\mu_{12}^2 E_1^2}{2\hbar^2} + i S_- (\omega_- - \omega_0) \right) [\gamma_1(S_-) + i \gamma_2(S_-)] + \frac{\mu_{12}^3 E_1^2 E_2}{4\hbar^3} \left(\frac{\omega_- - \omega_0 - i S_-}{\Delta_-} \right) [(\xi_2 - \xi_1) + i(\epsilon_1 + \epsilon_2)],$$

where

$$\gamma_1(S) = -\frac{\mu_{12} E_2}{\hbar} \left[\left(\frac{\mu_{12}^2 E_1^2}{\hbar^2 \Omega^2} - 1 \right) A_1 - \frac{\mu_{12}^2 E_1^2}{\hbar^2 \Omega^2} \left(\frac{A_2 - A_3}{2} \right) \right],$$

$$\gamma_2(S) = +\frac{\mu_{12} E_2}{\hbar} \left[\left(\frac{\mu_{12}^2 E_1^2}{\hbar^2 \Omega^2} - 1 \right) A - \frac{\mu_{12}^2 E_1^2}{\hbar^2 \Omega^2} \left(\frac{A_5 + A_6}{2} \right) \right],$$

$$A_1 = \frac{\alpha}{S^2 + \alpha^2}, \quad A_2 = \frac{\alpha + \Omega}{S^2 + (\alpha + \Omega)^2}, \quad A_3 = \frac{\Omega - \alpha}{S^2 + (\alpha - \Omega)^2}, \quad A_4 = \frac{S}{S^2 + \alpha^2}, \quad A_5 = \frac{S}{S^2 + (\alpha - \Omega)^2}, \quad A_6 = \frac{S}{S^2 + (\alpha + \Omega)^2},$$

and where

$$\xi_2 = \left(\frac{\omega_- - \omega_0}{\Omega^2} \right) \left[\frac{S}{S^2 + \alpha^2} - \frac{1}{2} \left(\frac{S}{S^2 + (\alpha - \Omega)^2} + \frac{S}{S^2 + (\alpha + \Omega)^2} \right) \right] + \frac{1}{2\Omega} \left(\frac{S}{S^2 + (\alpha - \Omega)^2} - \frac{S}{S^2 + (\alpha + \Omega)^2} \right),$$

$$\xi_1 = \frac{1}{2\Omega} \left(\frac{S}{S^2 + (\alpha - \Omega)^2} - \frac{S}{S^2 + (\alpha + \Omega)^2} \right) - \left(\frac{\omega_- - \omega_0}{\Omega^2} \right) \left[\frac{S}{S^2 + \alpha^2} + \frac{1}{2} \left(\frac{S}{S^2 + (\alpha - \Omega)^2} + \frac{S}{S^2 + (\alpha + \Omega)^2} \right) \right],$$

$$\epsilon_1 = \frac{1}{2\Omega} \left[\frac{\alpha + \Omega}{S^2 + (\alpha + \Omega)^2} + \frac{\Omega - \alpha}{S^2 + (\alpha - \Omega)^2} \right] + \left(\frac{\omega_- - \omega_0}{\Omega^2} \right) \left[-\frac{\alpha}{S^2 + \alpha^2} + \frac{1}{2} \left(\frac{\alpha + \Omega}{S^2 + (\alpha + \Omega)^2} - \frac{\Omega - \alpha}{S^2 + (\alpha - \Omega)^2} \right) \right],$$

$$\epsilon_2 = \left(\frac{\omega_- - \omega_0}{\Omega^2} \right) \left[-\frac{\alpha}{S^2 + \alpha^2} + \frac{1}{2} \left(\frac{\alpha + \Omega}{S^2 + (\alpha + \Omega)^2} - \frac{\Omega - \alpha}{S^2 + (\alpha - \Omega)^2} \right) \right] + \frac{1}{2\Omega} \left[\frac{\Omega + \alpha}{S^2 + (\alpha + \Omega)^2} + \frac{\Omega - \alpha}{S^2 + (\alpha - \Omega)^2} \right].$$

Thus

$$\left(\frac{dW}{dt}\right)_H = -N \frac{\hbar \omega_0}{2} \frac{\mu_{12} E_2}{\hbar \tau} \text{Re} \langle Z_1 \rangle_{\phi_0, \phi_1},$$

which gives Eq. (2.23).

$$\alpha = \omega_+ - \omega_-, \quad \phi_1 = \alpha t' + \phi_0, \quad \phi_0 = k z_0.$$

Thus

$$\Gamma_1 - \Gamma_2 = \frac{1}{2} e^{i\phi_1} [\delta r_2'(S_-) + i \delta r_1'(S_-)] + \text{c.c.},$$

where

$$S_- = 1/\tau - i\alpha, \quad S_+ = S_-^* = 1/\tau + i\alpha.$$

Replacing $2(\Gamma_1 - \Gamma_2)$ by Z_1 and averaging over the phases ϕ_0 and ϕ_1 , we get

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† Present address: Instituto de Física Universidade Estadual de Campinas, C. P. 1170, Campinas, São Paulo, Brasil.

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