Effect of velocity-changing collisions upon optical coherences in a three-level system

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In this work, a theory of laser nonlinear spectroscopy and its application to the study of atomic collisions is developed. A three-level system (TLS) is considered in low-pressure gases. The effect of weak velocity-changing collisions (vcc) upon atomic coherences is introduced into the TLS equations established by Hänsch and Toschek. It appears that a TLS is suitable for the observation of the effect of vcc upon optical coherences.

I. INTRODUCTION

The fruitful development of nonlinear laser spectroscopic techniques during the last few years has provided new evidence for collision-induced homogeneous broadening, shifts, and even distortion of spectral lines in low-pressure gases. Saturated absorption[1-4] and two-photon[5] spectroscopy have been used for such studies.

The use of nonlinear spectroscopy to study collisional effects represents a marked improvement over linear spectroscopy in measuring the effects of velocity-changing collisions (vcc). In linear spectroscopy, one starts with a thermalized sample. Consequently, elastic collisions do not alter the velocity distribution of the various level populations of the atom, which are already in equilibrium. Collisions will affect only the coherence or off-diagonal density-matrix elements, leading to a disturbance of the phase of the atomic oscillators. In general this phase disturbance consists of an inseparable combination of velocity-changing and phase-interrupting effects, although, in certain limits (see below), one contribution can dominate. In linear spectroscopy, effects of vcc are easily lost in the large widths of spectral profiles arising from the Doppler effect. In some cases, the effect of vcc on atomic coherences can be detected by a narrowing of the Doppler profile, but, in general, vcc are difficult to detect in linear spectroscopy.

Nonlinear Doppler-free spectroscopy provides more promise for studying subtle collisional processes. In such experiments one excites a given velocity group of atoms. By probing the system, one can determine the rethermalization of this population velocity group resulting from collisions. Moreover, there are systems where one can attempt to study the effect of velocity-changing collisions on the atomic coherences.

In a three-level system, it is possible to have a saturated absorption signal arising solely from terms related to optical coherences and not dependent on populations. By examining the effect of collisions on these profiles, one eliminates any problems arising from velocity-changing collisions on populations which would tend to mask their effects on "coherences."

There exists a large literature of calculations of velocity-changing collision effects in both linear and nonlinear spectroscopy.[6-10] To our knowledge, no calculation exists in which the model of weak velocity-changing collisions affecting internal coherences in a three-level system (TLS) has been fully explored. In this work, such a model is adopted and may prove relevant to explain recent experimental results involving saturation spectroscopy in a three-level system.[11]

II. TLS UNIDIRECTIONAL SPECTROSCOPY

In a typical experiment, two cw monochromatic collinear laser beams interact with an atomic or molecular gas contained in a low-pressure cell (Fig. 1). The field $E(\omega, k)$ is resonant with and saturates the 1-2 transition alone, selectively exciting optical dipoles with an axial velocity close to zero. The field $E'(\omega', k')$ with a frequency ω' close to that of the 2-3 transition monitors the changes in the velocity distribution and phase coherences of the system, induced by the pump field in the presence or absence of collisions.

The parameter measured in typical experiments[11] is that part of the absorption coefficient of the probe beam containing saturation effects; the most interesting situation occurs with unidirectional beams and $k > k'$.

A theoretical description of this system was given by Hänsch and Toschek.[12] Using perturbation theory, neglecting collisions, and assuming

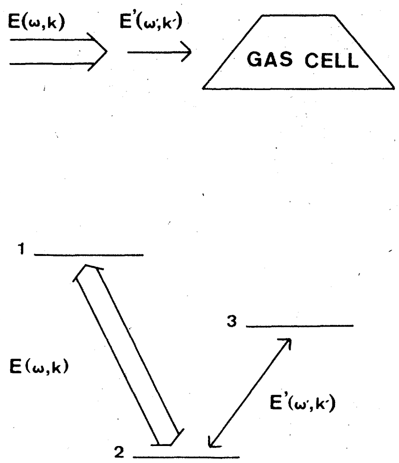

FIG. 1. Experimental situation and the energy-level scheme considered in this work. The frequencies of the fields E and E' are denoted by ω and ω', and the magnitudes of the propagation vectors by k and k'.

the Doppler limit (i.e., Doppler width much larger than decay rates), they obtained the term proportional to E^2 in the absorption coefficient for the field E' as

$$\alpha_3 = 4\pi k' E^2 \operatorname{Im}\chi_3,$$

where

$$\operatorname{Im}\chi_3 = 2\pi^{1/2}\hbar^{-3}\mu_{12}^2\mu_{23}^2(k\bar{v})^{-1}\exp\left(-\frac{(\omega-\omega_{12})^2}{(k\bar{v})^2}\right)$$
$$\times\left[N_{21}\left(\frac{1}{\gamma_2\Gamma_B^0}\,\frac{1}{1+Y^2}+\frac{\omega_{23}}{\omega_{12}}\,\frac{1}{\Gamma_B^0\Gamma_N^0}\,\frac{1-XY}{(1+X^2)(1+Y^2)}\right)\right.$$
$$\left.+N_{23}\left(1-\frac{\omega_{23}}{\omega_{12}}\right)\frac{1}{(\Gamma_N^0)^2}\,\frac{1-X^2}{(1+X^2)^2}\right], \tag{1}$$

N_{ij} is the population difference between i and j levels at equilibrium in the absence of all fields, γ_i is a level decay rate, γ_{ij} is the natural decay rate of the i–j coherence, ω_{ij} is a resonance frequency, μ_{ij} is the electric dipole moment of the i-j transition, $\bar{v}$ is the most probable atomic speed, and

$$\Gamma_N^0 = (1-\omega_{23}/\omega_{12})\gamma_{23} + (\omega_{23}/\omega_{12})\gamma_{13},$$
$$\Gamma_B^0 = \gamma_{23} + (\omega_{23}/\omega_{12})\gamma_{12},$$
$$X\Gamma_N^0 = Y\Gamma_B^0 = \omega' - \omega_{23} - (k'/k)(\omega-\omega_{12}).$$

Despite the apparent complexity of the expression for $\operatorname{Im}\chi_3$, it is possible to elucidate its physical content. The first term in square brackets is recognized as the usual saturation term resulting from the stepwise absorption of the fields E and E'. The second term is related with a two-quanta Raman-type transition connecting levels 1 and 3. The third term is another two-quanta contribution; it is noticeable that this term exists, even when the medium is transparent to the saturating field ($N_{21}=0$) contrary to the other two terms.

The third term corresponds, in atomic density-matrix formalism, to the following perturbative path:

$$\begin{Bmatrix}\rho_{22}^{(0)}\\ \rho_{33}^{(0)}\end{Bmatrix} \xrightarrow{E'} \rho_{23}^{(1)} \xrightarrow{E} \rho_{13}^{(2)} \xrightarrow{E} \rho_{23}^{(3)}.$$

This path does not depend upon saturated diagonal matrix elements. Therefore, vcc effects on populations play no role in this chain, making it well suited for studying the effect of collisions on optical coherences. Consequently, an experimental situation of interest occurs when the population inversion N_{21}, prepared by suitable pumping procedure, is much smaller than N_{23}. In this paper we are concerned only with third term of (1).

The steady-state equations of motion giving rise to the contribution of the third term in (1) are given by

$$[\gamma_{23} - i(\Delta' - k'v_z)]\tilde{\rho}_{23}^{(1)}(\vec{v}) = -i\beta'[\rho_{22}^{(0)}(\vec{v}) - \rho_{33}^{(0)}(\vec{v})],$$
$$\{\gamma_{13} + i[\Delta - \Delta' - (k-k')v_z]\}\tilde{\rho}_{13}^{(2)}(\vec{v}) = i\beta\tilde{\rho}_{23}^{(1)}(\vec{v}), \tag{2}$$
$$[\gamma_{23} - i(\Delta' - k'v_z)]\tilde{\rho}_{23}^{(3)}(\vec{v}) = i\beta\tilde{\rho}_{13}^{(2)}(\vec{v}),$$

where the pump and probe detunings are $\Delta = \omega - \omega_{12}$ and $\Delta' = \omega' - \omega_{23}$, respectively, $\tilde{\rho}_{ij}(\vec{v})$ is a density-matrix element in a radiation interaction representation,

$$\tilde{\rho}_{13}(\vec{v}) = \rho_{13}(\vec{v}, z, t)\exp\{i[(k-k')z + (\omega'-\omega)t]\},$$
$$\tilde{\rho}_{23}(\vec{v}) = \rho_{23}(\vec{v}, z, t)\exp[i(\omega' t - k'z)],$$

and β and β' are the Rabi frequencies of the transitions and are assumed to be much smaller than γ_{ij} in order that the perturbation calculation be valid.

III. THE EFFECT OF ELASTIC COLLISIONS UPON OPTICAL COHERENCES: A REASONABLE MODEL

The problem of interaction with an electromagnetic field of atoms or molecules subjected to collisions in a gas has already given rise to considerable theoretical studies (a comprehensive bibliography can be found in Ref. 9). The dynamical as well as the internal aspects of collisions have been extensively investigated and totally quantum-mechanical treatments have been achieved.[7,8] However, a lack of information on the interatomic potential prevents one from determining the scattering amplitudes which play a prominent role in all these theoretical calculations. Especially, the angular dependence of these amplitudes seems to be very sensitive—at least in the medium-angle diffusion region—to the precise shape of the potential.

As a consequence, we shall choose a simple empirical model to take into account the effect of elastic collisions upon optical coherences (off-diagonal density-matrix elements).

We assume a separation of collisions into two groups:

(i) Those which occur in a "near region," where the interaction potentials of the two levels involved in the transition are very different. These collisions result in a large abrupt change in phase for the off-diagonal density-matrix elements and consequently in a destroying of the coherence. In other words, coherence cannot be drifted by such a collision from one velocity class to another.

(ii) Those which occur in a "far region," where the interaction potentials of the two levels are so close that the phase-interrupting effect is negligible These collisions result only in a change in the velocity associated with the atomic dipole. This change is necessarily small due to the size of the impact parameter but may still be detectable by laser-spectroscopy techniques. A consequence of these assumptions is a statistical independence of coherence-destroying and velocity-changing collisions. The small change in the velocity of the atom, enables us to use the so-called weak-collision approximation.

The model is expressed mathematically by the addition of a term $R_{ij}|_{\rm coll}$ to the right-hand side of (2):

$$R_{ij}|_{\rm coll} = -\gamma_{ij}^{\rm ph}(\vec{v})\tilde{\rho}_{ij}(\vec{v}) - \Gamma_{ij}^{\rm vc}(\vec{v})\tilde{\rho}_{ij}(\vec{v}) + \int W_{ij}(\vec{v}\,' \to \vec{v})\tilde{\rho}_{ij}(\vec{v}\,')\, d\vec{v}\,', \qquad (3)$$

where $\gamma_{ij}^{\rm ph}$ is the rate of phase-interrupting collisions, $W_{ij}(\vec{v}\,' \to \vec{v})$ is the collision kernel for vcc, and

$$\Gamma_{ij}^{\rm vc}(\vec{v}) = \int d\vec{v}\,'\, W_{ij}(\vec{v} \to \vec{v}\,')$$

is the rate of vcc. The collision kernel is assumed to be important in the region where $|\vec{v}\,' - \vec{v}|$ is much smaller than the mean atomic velocity.

IV. SOLUTION OF THE EQUATIONS OF MOTION

First, we can reduce the velocity dependence of the problem to one dimension by noticing that the orthogonal velocity distribution of the active atoms is hardly perturbed by the saturating beam (propagating along v_z). Therefore one can factorize density-matrix elements:

$$\tilde{\rho}_{ij}(\vec{v}) = \underline{\tilde{\rho}}_{ij}(v_z) W_M(\vec{v}_\perp),$$

where $W_M(\vec{v}_\perp)$ is the equilibrium velocity distribution for the transverse velocity components.

Including the collision terms in equation set (2) and summing it over $\vec{v}_\perp$, neglecting the velocity dependence of $\Gamma^{\rm vc}$ and $\gamma^{\rm ph}$, one gets

$$\begin{aligned}
&[\Gamma_{23} - i(\Delta' - k'v_z)]\underline{\tilde{\rho}}_{23}^{(1)}(v_z) = -i\beta' N_{23}(v_z) + \int W_{23}(v_z', v_z)\underline{\tilde{\rho}}_{23}^{(1)}(v_z')\, dv_z',\\
&\{\Gamma_{13} + i[\Delta - \Delta' - (k-k')v_z]\}\underline{\tilde{\rho}}_{13}^{(2)}(v_z) = i\beta\underline{\tilde{\rho}}_{23}^{(1)} + \int W_{13}(v_z', v_z)\underline{\tilde{\rho}}_{13}^{(2)}(v_z')\, dv_z',\\
&[\Gamma_{23} - i(\Delta' - k'v_z)]\underline{\tilde{\rho}}_{23}^{(3)}(v_z) = i\beta\underline{\tilde{\rho}}_{13}^{(2)} + \int W_{23}(v_z', v_z)\underline{\tilde{\rho}}_{23}^{(3)}(v_z')\, dv_z'
\end{aligned} \qquad (4)$$

where

$$\Gamma_{ij} = \gamma_{ij} + \gamma_{ij}^{\rm ph} + \Gamma_{ij}^{\rm vc},$$

$$W_{ij}(v_z', v_z) = \int d\vec{v}_\perp\, d\vec{v}_\perp'\, W_M(\vec{v}_\perp')W_{ij}(\vec{v}\,' \to \vec{v}),$$

and

$$N_{23}(v_z) = N_{23} W_M(v_z).$$

For weak collisions and low pressures, the collision kernels in (4) can be chosen to depend solely on the difference $|v_z - v_z'|$. While kernels of this nature do not satisfy detailed balancing and cannot give rise to collisional narrowing of spectral profiles, they may be used without significant error provided that the effective collisional mean free path is large compared with the appropriate wavelength in the problem. Specifically, one requires[13]

$$k\bar{v} \gg \Gamma_{23}\tilde{u}_{23}^2/\bar{v}^2,$$

$$|k - k'|\bar{v} \gg \Gamma_{13}\tilde{u}_{13}^2/\bar{v}^2,$$

where $\tilde{u}_{ij}$ is the width of the kernel $W_{ij}(v_z - v_z')$ and $\Gamma_{ij}\tilde{u}_{ij}^2/\bar{v}^2$ is the effective[13] collision rate. In the frame of this approximation, the following equality holds:

$$\Gamma_{ij}^{\rm vc} = \int_{-\infty}^{+\infty} W_{ij}(v_z, v_z')\, dv_z = \int_{-\infty}^{+\infty} W_{ij}(v_z', v_z)\, dv_z'. \qquad (5)$$

In what follows, we write

$$W_{ij}(|v_z - v_{z'}|) \equiv W_{ij}(v).$$

The equations of motion are most easily solved by a Fourier-transform method.[7] The Fourier transform $\mathcal{F}(\tau)$ of an arbitrary function $f(v_z)$ is defined as

$$\mathcal{F}(\tau) = \left(\frac{k'}{2\pi}\right)^{1/2} \int_{-\infty}^{+\infty} \exp(ik'v_z\tau) f(v_z)\, dv_z .$$

Equation set (4) may be transformed into

$$[\Gamma_{23}-\mathcal{W}'_{23}(\tau)-i\Delta']\tilde{\underline{\mathcal{P}}}^{(1)}_{23}(\tau)+\frac{d\tilde{\underline{\mathcal{P}}}^{(1)}_{23}(\tau)}{d\tau}=i\beta'\mathfrak{N}_{23}(\tau)\,,$$

$$[\Gamma_{13}-\mathcal{W}'_{13}(\tau)+i(\Delta-\Delta')]\tilde{\underline{\mathcal{P}}}^{(2)}_{13}(\tau)-\frac{k-k'}{k'}\frac{d\tilde{\underline{\mathcal{P}}}^{(2)}_{13}(\tau)}{d\tau}=i\beta\tilde{\underline{\mathcal{P}}}^{(1)}_{23}(\tau)\,, \quad (6)$$

$$[\Gamma_{23}-\mathcal{W}'_{23}(\tau)-i\Delta']\tilde{\underline{\mathcal{P}}}^{(3)}_{23}(\tau)+\frac{d\tilde{\underline{\mathcal{P}}}^{(3)}_{23}(\tau)}{d\tau}=i\beta\tilde{\underline{\mathcal{P}}}^{(2)}_{13}(\tau)\,.$$

where $\mathfrak{N}_{23}$, $\tilde{\underline{\mathcal{P}}}_{ij}$, and $\mathcal{W}$ are the Fourier transforms of N_{23}, $\underline{P}_{ij}$, and W_{ij}, with

$$\mathcal{W}'_{ij}(\tau)=(2\pi/k')^{1/2}\mathcal{W}_{ij}(\tau)\,.$$

This system is solved as follows:

$$\tilde{\underline{\mathcal{P}}}^{(1)}_{23}(\tau)=-i\beta'\int_{-\infty}^{\tau}\mathfrak{N}_{23}(\tau')\times\exp\left(\int_{\tau}^{\tau'}[-i\Delta'+\Gamma_{23}-\mathcal{W}'_{23}(\tau'')]\,d\tau''\right)d\tau'\,, \quad (7)$$

$$\tilde{\underline{\mathcal{P}}}^{(2)}_{13}(\tau)=i\frac{k'}{k-k'}\beta\int_{\tau}^{\infty}d\tau'\tilde{\underline{\mathcal{P}}}^{(1)}_{23}(\tau')\exp\left(-\int_{\tau}^{\tau'}d\tau''\frac{k'}{k-k'}[i(\Delta-\Delta')+\Gamma_{13}-\mathcal{W}'_{13}(\tau'')]\right), \quad (8)$$

$$\tilde{\underline{\mathcal{P}}}^{(3)}_{23}(\tau)=i\beta\int_{-\infty}^{\tau}\tilde{\underline{\mathcal{P}}}^{(2)}_{13}(\tau')\exp\left(\int_{\tau}^{\tau'}d\tau''[-i\Delta'+\Gamma_{23}-\mathcal{W}'_{23}(\tau'')]\right)d\tau'\,. \quad (9)$$

Each of the exponential factors acts as a propagator containing collision and natural damping. Each one drives a density-matrix element from its creation at time τ' due to a field interaction, up to time τ.

Bringing (7) into (8), and (8) into (9), one obtains

$$\tilde{\underline{\mathcal{P}}}^{(3)}_{23}(\tau)=-i\beta^2\beta'\frac{k'}{k-k'}\int_{-\infty}^{\tau}d\tau'\int_{\tau'}^{\infty}d\tau''\int_{-\infty}^{\tau''}d\tau'''\,\mathfrak{N}_{23}(\tau''')\times\exp\Big((-i\Delta'+\Gamma_{23})(\tau'-\tau)-\tilde{\mathcal{W}}_{23}(\tau')+\tilde{\mathcal{W}}_{23}(\tau)-\frac{k'}{k-k'}[i(\Delta-\Delta')+\Gamma_{13}](\tau''-\tau')$$
$$+\frac{k'}{k-k'}[\tilde{\mathcal{W}}_{13}(\tau'')-\tilde{\mathcal{W}}_{13}(\tau')]+(-i\Delta'+\Gamma_{23})(\tau'''-\tau'')-\tilde{\mathcal{W}}_{23}(\tau''')+\tilde{\mathcal{W}}_{23}(\tau'')\Big)\,, \quad (10)$$

with

$$\tilde{\mathcal{W}}_{ij}(\tau)=\int_0^{\tau}d\tau'\mathcal{W}'_{ij}(\tau')=\int_{-\infty}^{+\infty}dv_z\frac{\sin(k'v_z\tau)}{k'v_z}W_{ij}(v_z)\,, \quad (11)$$

where we have used the fact that $W_{ij}(v_z)$ is an even function.

We are interested in the integral of $\tilde{\underline{\rho}}^{(3)}_{23}(v_z)$ over velocity, since

$$E^2E'\,\mathrm{Im}\chi_3=\mu_{23}\,\mathrm{Im}\int_{-\infty}^{+\infty}dv_z\tilde{\underline{\rho}}^{(3)}_{23}(v_z)\,. \quad (12)$$

The transformation back to velocity space is performed using

$$\int_{-\infty}^{+\infty}dv_z\tilde{\underline{\rho}}^{(3)}_{23}(v_z)=\left(\frac{2\pi}{k'}\right)^{1/2}\tilde{\underline{\mathcal{P}}}^{(3)}_{23}(0)\,. \quad (13)$$

Additional approximations provide a more tractable expression. First, as has been done in arriving at (1), we may assume that the Doppler limit is fulfilled. Consequently, we approximate $N_{23}(v_z)$ by $N_{23}(\Delta/k)$ leading to a value

$$\mathfrak{N}_{23}(\tau)=(2\pi/k')^{1/2}N_{23}(\Delta/k)\delta(\tau)$$

and the result

$$\int_{-\infty}^{+\infty}\tilde{\underline{\rho}}^{(3)}_{23}(v_z)dv_z=i\beta^2\beta'\frac{k'}{k-k'}N_{23}\left(\frac{\Delta}{k}\right)\frac{2\pi}{k'}\left[\int_0^{\infty}d\tau\exp\left((i\Omega'-\Gamma_N)\frac{k}{k-k'}\tau+\frac{k'}{k-k'}\tilde{\mathcal{W}}_{13}(\tau)+\tilde{\mathcal{W}}_{23}(\tau)\right)\right]^2, \quad (14)$$

where

$$\Gamma_N=(1-\omega_{23}/\omega_{12})\Gamma_{23}+(\omega_{23}/\omega_{12})\Gamma_{13}$$

and

$$\Omega'=\omega'-\omega_{23}-(k'/k)(\omega-\omega_{12})\,.$$

To go further, one needs an explicit form for the collision kernel. Keeping in mind that we have assumed that only distant collisions contribute to the velocity-changing collision kernel, we may conclude that the semiclassical approximation conditions are fulfilled in that region. Then, a Gaussian kernel can be obtained as has been shown by Kolchenko *et al.*[14]

Taking

$$W_{ij}(v_z)=(\Gamma_{ij}^{vc}/\sqrt{\pi}\,\tilde{u}_{ij})\exp(v_z^2/\tilde{u}_{ij}^2)\,,$$

using (10), and doing a little algebra, one finally gets

$$\tilde{W}_{ij}(\tau)=\sqrt{\pi}\,\frac{\Gamma_{ij}^{vc}}{k'\tilde{u}_{ij}}\,E_2\!\left(\frac{k'\tilde{u}_{ij}\tau}{2}\right),$$

where $E_2(x)$ stands for the error function,

$$E_2(x)=\frac{2}{\sqrt{\pi}}\int_0^x e^{-t^2}dt\,.$$

The resulting contribution to the imaginary part of χ_3 is now in a form well suited for a computer calculation:

$$\mathrm{Im}\chi_3=\frac{2\pi}{k}\left(1-\frac{\omega_{23}}{\omega_{12}}\right)\hbar^{-3}\mu_{12}^2\mu_{23}^2N_{23}\left(\frac{\Delta}{k}\right)$$
$$\times\,\mathrm{Re}\left\{\int_0^\infty d\tau\,\exp\left[(i\Omega'-\Gamma_N)\tau+\frac{\omega_{23}}{\omega_{12}}\sqrt{\pi}\,\frac{\Gamma_{13}^{vc}}{k'\tilde{u}_{13}}E_2\!\left(\frac{k'\tilde{u}_{13}\tau}{2}\right)+\left(1-\frac{\omega_{23}}{\omega_{12}}\right)\sqrt{\pi}\,\frac{\Gamma_{23}^{vc}}{k'\tilde{u}_{23}}E_2\!\left(\frac{k'\tilde{u}_{23}\tau}{2}\right)\right]\right\}^2. \qquad (15)$$

This is the final expression that we obtain in this paper, taking into account the effect of vcc. When $\Gamma_{13}^{vc}=\Gamma_{23}^{vc}=0$, one can easily identify (15) with the third term in (1).

V. DISCUSSION

In this section, we emphasize that the effect of vcc depends strongly upon the size of the width of the collision kernel relative to the atomic decay rates. In a characteristic situation, the shape of the signal profile may be noticeably distorted due to the effect of vcc.

In order to isolate the effect of vcc in a single term in Eq. (14), we make use of (5) and obtain an alternative form for (14):

$$\int_{-\infty}^{+\infty}\tilde{\rho}_{23}^{(3)}(v_z)\,dv_z=i\beta^2\beta'\frac{k'}{k-k'}N_{23}\left(\frac{\Delta'}{k'}\right)\frac{2\pi}{k}\left[\int_0^\infty d\tau\,\exp\left(i\Omega'-\Gamma_N^0-\Gamma_N^{ph}\right)\frac{k}{k-k'}\tau+\frac{k}{k-k'}V(\tau)\right)\Big]^2, \qquad (16)$$

where

$$V(\tau)=\int_{-\infty}^{\infty}dv_z\left(\frac{\sin(k'v_z\tau)}{k'v_z}-\tau\right)W_N(v_z) \qquad (17)$$

and

$$W_N(v_z)=(k'/k)W_{13}(v_z)+[(k-k')/k]W_{23}(v_z)\,.$$

The entire effect of vcc is contained in $V(\tau)$. To analyze (16) and (17), a parameter of physical interest is the coherence time Γ_c^{-1}, the time at which the real part of the exponential argument in (16) is unity. Depending on the relative size of $k\tilde{u}$ and Γ_c, two extreme situations occur [$\tilde{u}^2$ is a mean-square change of velocity during a collision associated with the kernel $W_N(v_z)$].

(i) $\Gamma_c\gg k\tilde{u}$. In this limit, the sine function in (17) can be expanded in a power series, and the first nonvanishing contribution leads to an exponential in (16) of the form[15]

$$\exp[i(\Omega'-\Gamma_N^0-\Gamma_N^{ph})\frac{k}{k-k'}\tau-\frac{k}{k-k'}\frac{\Gamma_N^{vc}}{6}\tau(k'\tilde{u}\tau)^2]\,.$$

The presence of a cubic term in τ indicates that vcc can give rise to line shape different in form from that obtained assuming either no collisions or only phase-interrupting collisions. The coherence time Γ_c^{-1} is the solution of the cubic equation

$$(\Gamma_N^0+\Gamma_N^{ph})/\Gamma_c+\tfrac{1}{6}\,\Gamma_N^{vc}(k'\tilde{u})^2/\Gamma_c^3=1\,.$$

The relative values of $k\tilde{u}$, $\Gamma_N^0+\Gamma_N^{ph}$, Γ_N^{vc} determine the specific effect of the vcc.

For example, if $\Gamma_N^{vc}(k'\tilde{u})^2\ll(\Gamma_N^0+\Gamma_N^{ph})^3$, the vcc will not significantly alter the phase of the off-diagonal density matrix elements during the coherence time $\Gamma_c^{-1}\simeq(\Gamma_N^0+\Gamma_N^{ph})^{-1}$. On the other hand, if $\Gamma_N^{vc}(k'\tilde{u})^2\gg(\Gamma_N^0+\Gamma_N^{ph})^3$, it is the vcc which effectively determine coherence time $\Gamma_c^{-1}\simeq[\Gamma_N^{vc}(k'\tilde{u})^2]^{-1/3}$. In this case, the vcc lead to a change in the functional form of the line shape resulting from phase excursions caused by a number of weak vcc with $k\tilde{u}<\Gamma_c$.

(ii) $\Gamma_c\ll k'\tilde{u}$. The sine function in (17) is rapidly varying for all τ of interest and may be neglected leading to an exponential in (16) of the form

$$\exp[i(\Omega'-\Gamma_N^0-\Gamma_N^{ph}-\Gamma_N^{vc})\tau k/(k-k')]\,.$$

The net effect of the vcc in this case is to add an additional rate Γ_N^{vc} to the destruction of atomic coherence. The inverse coherence time is now

$$\Gamma_c = \Gamma_N^0 + \Gamma_N^{ph} + \Gamma_N^{vc} .$$

Therefore, this situation occurs when $k'\bar{u} \gg \Gamma_N^0 + \Gamma_N^{ph} + \Gamma_N^{vc}$ so that each collision is strong enough to cause a total phase destruction of the coherences.

When none of the above limits are realized, (15) and (17) should be used. To have an insight into the physical meaning of these results it is useful to remember the case of level populations (i.e., diagonal matrix elements) on which the effects of vcc is simply understood as a redistribution of the atoms over the velocity space, approaching thermal equilibrium. The vcc act in a pure-classical kinetic way and do not affect the total population of a level (integrated over velocity).

On the contrary, a change of velocity induces a change in the translation or Doppler phase of the off-diagonal density matrix element, which results in an additional decay term of this matrix element.[13] Consequently, the vcc affect the total atomic coherence between two levels (i.e., the coherence integrated over the velocity) and, at the same time, the signal profile.

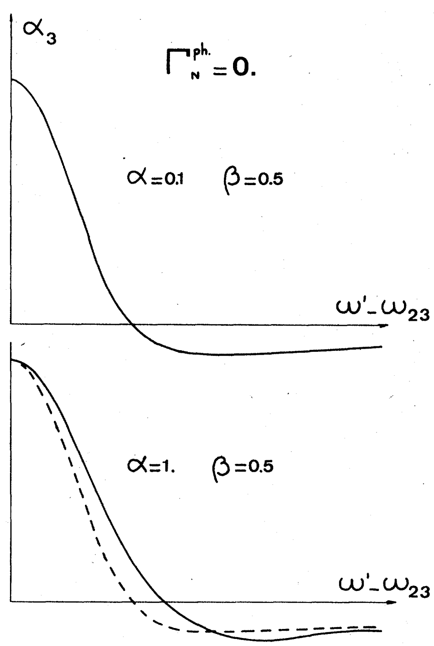

FIG. 2. Saturated absorption profiles for two values of the vcc kernel width. The dashed line represents α_3 in the absence of collisions, and the solid line represents it in the presence of vcc. The parameter α denotes the ratio of the kernel widths of Γ_N^0/k'. In this figure one assumes that there is no effect of phase-interrupting collisions.

We have illustrated these results of Figs. 2 and 3, where Γ^{ph} and N_{12} are fixed to zero, and the following parameters are used:

$$\alpha = k'\bar{u}/2\Gamma_N^0 , \quad \beta = \Gamma_N^{vc}/\Gamma_N^0 .$$

Dashed lines represent the absorption coefficient α_3 in the absence of collisions, and solid lines represent it in the presence of vcc.

Figure 2 shows the evolution of the profile with increasing kernel width, from $k'\bar{u} \ll \Gamma_N^0$ to $k'\bar{u} \sim \Gamma_N^0$. In Fig. 3, this width is constant and two different vcc rates are used. In the absence of vcc the ratio $\alpha_{3\,max}/\alpha_{3\,min}$ is a constant. In the presence of vcc, in addition to a broadening of the curve, the most interesting feature is a change in the value of $\alpha_{3\,max}/\alpha_{3\,min}$, providing a unique signature for the presence of vcc on optical coherences.

These results could be considered as complementary to those of Barantsov *et al.*[16] who have already pointed out the interest of observing the effect of the vcc upon the interference terms in a three-level system; they focused their calculations upon the study of the Dicke effect associated with a microwave transition between two levels connected to a third one by an optical transition and used a strong collision model.

Laser spectroscopy of molecular two-level systems (saturated absorption[17] and photon echo[13])

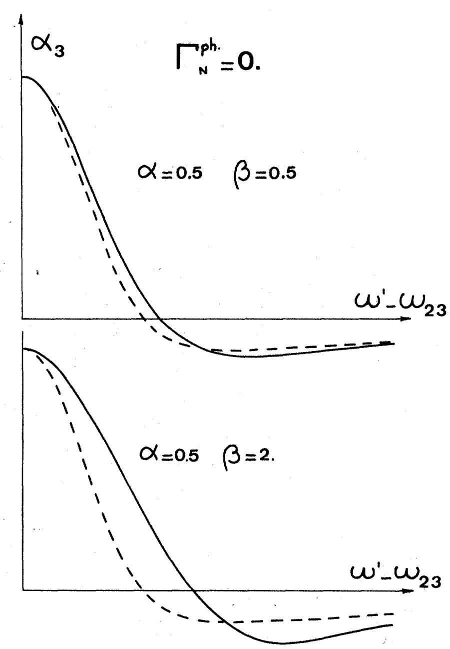

FIG. 3. Saturated absorption profiles for two values of the vcc rate. The parameter β denotes the ratio of the vcc rate to the decay rate Γ_N^0. The dashed and solid lines have the same meaning as in Fig. 2.

has already proved an efficient tool for observing the effect of vcc on optical coherences. On the other hand, observations of collision effects in a three-level system[18,19] have been consistent with an effect of vcc on level populations only; these studies have dealt mainly with the usual saturated contribution resulting from the stepwise absorption of the saturating and probe beams. In this paper, we have shown that TLS experiments, such as that referred to in Ref. 11, can also be used to search for the effect of vcc on optical coherences.

Note added in proof. An article[20] containing a similar calculational method for studying the effects of vcc on coherences using saturated absorption in a two-level system has recently appeared.

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$$E_{i3}(Z)=\int_0^\infty \exp(-t^3-3Zt)\,dt,$$

which, for any complex value of Z is represented by the series

$$E_{i3}(Z)=\frac{\pi}{3}\,\frac{Z^2}{\cos(\pi/6)}\sum_{m=0}^{\infty}\frac{(-)^m Z^{3m}}{\Gamma(m+\frac{4}{3})\Gamma(m+\frac{5}{3})}+\frac{\pi}{3\sin(\pi/3)}\left(\sum_{m=0}^{\infty}\frac{(-)^m Z^{3m}}{m!\,\Gamma(m+\frac{2}{3})}-Z\sum_{m=0}^{\infty}\frac{(-)^m Z^{3m}}{m!\,\Gamma(m+\frac{4}{3})}\right)$$

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