Shift and broadening of saturated absorption resonances due to curvature of the laser wave fronts

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Using frequency offset-locked spectroscopy of methane at 3.39 μm we show that the saturated absorption peaks can be significantly blue or red shifted if the wave fronts of the laser beam are spherical in the gas cell. Good agreement with theoretical prediction is found. We discuss suitable conditions to avoid this geometry-induced shift in optical frequency standards.

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In precision experiments using frequency-controlled laser saturation spectroscopy, we have sometimes observed a small asymmetry in the line profile.[1,2] It seemed to arise from some uncontrolled experimental effect associated with the laser beam geometry, perhaps curvature of the wave fronts. The dominating influence of the laser beam geometry on the saturated absorption line shape stimulated development of a theory[3-5] including not only the Gaussian spatial amplitude dependence but also the phase variation of the electromagnetic field associated with the wave-front curvature. (Because of the finite laser beam aperture, diffraction leads to curvature in any finite length of absorbing material.) We found that theory indeed predicted asymmetric profiles when such curved wave fronts were considered. In this paper we present experimental results which clearly demonstrate these shifting effects and find quantitative agreement with the theoretical expectations.

To make a quantitative experimental study, one needs a stable offset frequency reference such as that provided in the frequency offset-locked laser spectrometer.[1,6] The large absorption cell was replaced by a 20-cm test cell that could be placed at will in a laser beam of well-defined geometry. The cell pressure of 3.0 mTorr gives rise to a pressure broadening of about 45 kHz HWHM. The beam was measured to be 4.7 mm at the $1/e$ radius, corresponding to a transit linewidth of 15 kHz and to a 40-m confocal parameter. In the first experiment the cell was placed in this "parallel" beam which was retroreflected back through the cell and ultimately into the detector. The retroreflector was formed by an antireflection-coated lens of focal length $f=120$ cm with a plane mirror at its focus. The retroreflector's focus was accurately adjusted by observation of residual interference fringes with the laser (with the YIG isolators slightly detuned). Figure 1(a) shows the derivative resonance signal obtained under these conditions of collimated and matched beams, with R designating the signal from double-passed return beam. The signal T was obtained with an auxiliary detector seeing the small transmitted fraction of the light incident on the mirror. It can be seen from Fig. 1(a) that the two line shapes are symmetrical and have the same center. Computer least-squares fits of these curves gave the same unshifted center within the 500-Hz standard deviation of this output parameter. The linewidths obtained were 71 and 74 kHz HWHM, almost agreeing within the expected error, and consistent with a saturation parameter of 1.46 calculated from the laser power of 1 mW.[5,7]

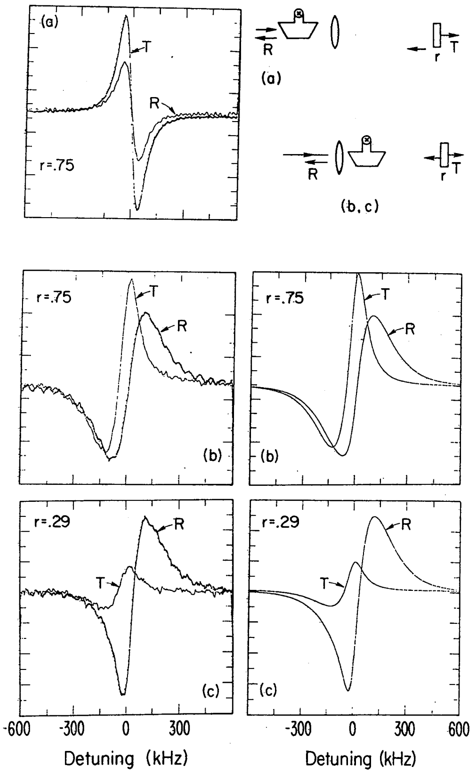

FIG. 1. Theoretical and experimental profiles. Methane pressure = 3.0 mTorr. Transmitted beam's signal T, double-passed signal R. (a) Parallel beam case $w_0 = 4.7$ mm. Upper sketch shows cell location in parallel beam. (b), (c) Curved wave-front cases, cell located inside retroreflector (lower sketch). The cell center is 12 units of the half-confocal parameter from the waist in the focal plane of the cat's eye. Experimental profiles on the left, theoretical profiles on the right. See text for details. (b) Reflectivity $r = 0.75$; (c) $r = 0.29$. (*Note*: The data sign reversal is not meaningful.)

Then the cell was removed from the parallel beams and, as indicated in Fig. 1, was placed inside the cat's eye, where the matched curved beams are characterized by the new mode waist $w_0 = 0.275$ mm (located at the focus). The center of the cell was 85 cm from this waist. In this location of curved wave fronts, derivative spectra were obtained as before for several values of the effective reflection coefficient r. For $r \simeq 100\%$ the reflected beam was centered and symmetric, although broadened. For lower reflectivities, dramatic shift, broadening, and asymmetry were observed. In Fig. 1(b) we show the cases of $r = 0.75$ and $r = 0.29$, with the signal of the purely converging transmitted probe beam denoted T as before.

The *converging* probe beam gives a *red-shifted* line as predicted by the theory. In Fig. 1(b), one notes that the zero of the first harmonic lock-in signal of the transmitted probe beam is shifted by -49 ± 3 kHz, the same in both cases. The peak-to-peak width, times $\sqrt{3}/2$, is 113 ± 5 kHz and is also the same in both cases. Neglecting higher-order saturation effects, the reflected signal R is the sum of the red-shifted asymmetric converging probe profile, weighted by the reflection coefficient, and of the blue-shifted signal arising from the diverging wave acting as the probe.[8] The result is a reduction of the asymmetry for the double-passed signal, with a net shift of $+15 \pm 4$ kHz for the $r = 0.75$ case and of $+41 \pm 4$ kHz for $r = 0.29$. To check the influence of saturation, the power of the input beam was attenuated by a factor of 1.6. No significant changes in the shifts or widths with power are observed, showing that the saturation parameter is reduced by the curvature of the wave fronts (since the cell is far from the focus).

In a paper describing the third-order theory[4] of saturated absorption spectroscopy, we obtained for each recoil peak the geometry-dependent part of the line shape for the probe beam. We found

$$gJ_\alpha = \tfrac{1}{2}\eta_\alpha \,\mathrm{Im}\int_0^\infty dt\{\exp[-2(\eta_{ab} - i\xi)t]/Y\} \times [\exp(X - iY)E_1(X - iY) - \exp(X + iY)E_1(X + iY)]S(2Mt),$$

where the decay rate in level $\alpha = a$, b is $\eta_\alpha = \gamma_\alpha T_{tr}$, and where the dipole decay rate is $\eta_{ab} = \gamma_{ab} T_{tr}$, both in units of the average reciprocal transit time at the beam waist, $T_{tr} = w_0/u$. The detuning parameter is $\xi = (\omega - \omega_0 \pm \hbar\omega^2/2Mc^2)T_{tr}$, where the $-$ sign for the recoil[9] term is to be associated with the lower state. E_1 is the exponential integral function,[10] and $X = \eta_\alpha\,(1 + is)t$, $Y = \eta_\alpha (1 + s^2)^{1/2}(1 + t^2 - 2i\delta_2 t)^{1/2}$, and $\delta_2 = (\omega_0 u^2/2c^2)T_{tr}$ is the second-order Doppler shift in units of the reciprocal transit time. The profile is given here only for the case of two matched Gaussian beams of confocal parameter b and waist radius w_0 counterpropagating along the z axis. The parameter $s = 2z/b$ measures the distance from the common waist. Inclusion of the function

$$S(2Mt) = 1$$
$$= 2i^n J_n(2Mt)$$

allows us to obtain either the nonderivative line shape or its nth Fourier component for lock-in detection.[11] M is the frequency modulation amplitude in units of T_{tr}^{-1}.

A Fortran program has been written to evaluate this profile using an adaptive "Simpson rule" technique. Appropriate rational approximations are used for $E_1(Z)$ and for the Bessel function $J_n(2Mt)$. For $s \neq 0$ the line profile is, of course, asymmetric, and one can interchange the role of probe and saturation beams by letting $s \to -s$. Reference 4 contains asymptotic formulas for the free-flight and collision-dominated regimes.

The formulas described above were evaluated for the conditions of the curved wave-front experiments ($\gamma/2\pi = 45$ kHz, $T_{tr} = 0.49$ μsec, $M = 0.031$, $s = 2z/b = 12$), giving rise to the profiles on the right side of Fig. 1(b). The excellent agreement between theoretical and experimental profiles leaves no doubt that the observed shift and asymmetry are due to the influence of wave-front curvature. We emphasize that the theoretical shift of -48.5 kHz for the single-pass line shape is in quantitative agreement with experiment, although the comparison contains no adjustable frequency-scaling parameter. The predicted shifts also agree in the two cases for the double-passed data (R), although the profile superposition argument given above makes this a weaker test of the theory. The success of these comparisons makes it attractive to consider a new series of experiments with still more precise definition of the beam geometry and collision broadening. The theory could then be refined to account for the finite length of the absorption cell. One can also use the high-field theory[3,5] to deal with higher-order saturation effects.

We have shown that the theory can be applied to obtain quantitative line profiles in saturated absorption spectroscopy. As one interesting example, we can estimate that in the typical He-Ne I_2-stabilized laser the calculated one-way shift of 2 kHz is reduced 100-fold by the effective reflection coefficient of 99%. However, in high-gain systems, appreciable shifts could result from use of a low reflectivity at the gas-cell end and from lack of symmetry.

Another problem with large path-integrated gains is the introduction of wave-front geometry changes through power- and frequency-dependent amplifier dispersion and radial gain distribution. Such geometry changes will then give rise to curvature-induced shifts. A natural direction away from these problems is the use of an expanded beam combined with a reasonable pressure (or natural damping) to suppress the influence of imperfect wavefronts. Such large confocal parameters also reduce the influence of the absorber gas lens self-focusing/self-defocusing effects. These conditions provide the further advantage of a power-stable and calculable second-order Doppler shift.[4,12] In addition, the interdependence of modulation amplitude and various shifts and asymmetries can be evaluated with the formula presented here.

To summarize, we find experimentally that strongly asymmetric profiles can indeed be produced in saturation spectroscopy when the molecules' free flight allows them to sample the wave-front curvature.[13] The described theory gives remarkably good agreement with the experiment.

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