Effect of field and transit broadening on the collisional displacement of an optical frequency standard

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(Submitted 22 February 1979)

Pis'ma Zh. Eksp. Teor. Fiz. **29**, No. 7, 428–432 (5 April 1979)

We show that the concurrent effect of the impact and field or impact and transit line broadening mechanisms leads to displacement of an optical frequency standard with variation in the field and transit line width of an isolated nonlinear resonance absorption of a molecular transition.

PACS numbers: 42.72. + h, 33.70.Jg, 33.35.Ex

1. Use of saturated absorption resonances of molecular gases as modern optical frequency standards permits highly accurate molecular transition frequency reproducibility (see, for example, Ref. 1). In recent experiments, the reproducibility accuracy depends almost entirely on the magnitude of the nonlinear absorption resonance with respect to the line center. The literature discusses how such a shift is affected by the presence of the hyperfine line structure,[2,3] quadratic Doppler effect,[3] recoil effect,[4] wavefront curvature[5] and a number of other factors.

In this paper we want to turn our attention to one more effect—leading to the resonance shift—which has not been discussed heretofore. This effect may be dominant in the case of an isolated transition in the absence of hyperfine structure.

2. The effect in question occurs as a result of a unique interaction of the collision line shift with other broadening mechanisms. In order to explain such an interaction, we shall recall the special features of collision broadening of nonlinear resonances at low pressures.

As we know, in those cases where the radiation transition line width is relatively small, the impact width and displacement of saturated absorption resonances of a molecular gas are nonlinear functions of density.[6,7] A qualitative explanation of such a dependence is as follows. The interval Δv of the velocity of molecules which contribute to the resonance projected on the direction of the field wave vector **k** is described by a ratio $|\Delta v| \lesssim \gamma/k$, where γ is the homogeneous linewidth. Thus, if collisions place the molecular velocity outside this interval—as is the case when condition $kv_0 \times \bar{\theta} \equiv \Delta\omega_D \bar{\theta} \gg \gamma$ is satisfied [where v_0 is mean-thermal molecular velocity, $\bar{\theta}$ is the mean scattering angle ($\bar{\theta} \ll 1$)]—the effect of the change in the molecule velocity in the presence of collisions contributes to line broadening and displacement. When the reverse inequality $\Delta\omega_D \bar{\theta} \ll \gamma$ is satisfied changes in the molecular velocity due to collisions cease to play the role of a broadening factor. In summary, the impact width and displacement of a line δ are described by different formulas for different values of γ[7]

$$\gamma + i\delta = \begin{cases} \Gamma + i\Delta + \nu\,, & \gamma << \Delta\omega_D \bar{\theta} \\ \Gamma + i\Delta\,, & \gamma >> \Delta\omega_D \bar{\theta} \end{cases}, \qquad (1)$$

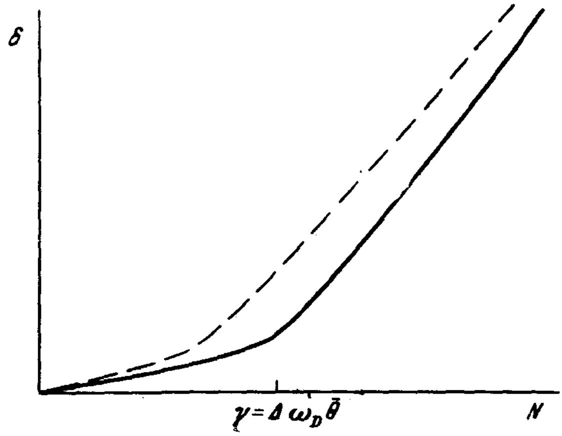

FIG. 1.

where Γ and Δ are line width and displacement in the conventional impact theory; the complex parameter $\nu = \nu' + i\nu''$ reflects the role of collisions with changing velocity. Γ, Δ and ν are proportional to the density of the disturbing gas N such that in a region of pressures, for which $\gamma = \Delta\omega_D\bar{\theta}$, the slopes of the curves $\delta(N)$ and $\gamma(N)$ change. Figure 1 shows the characteristic dependence of resonance displacement on pressure $\delta(N)$ for molecular vibrational-rotational transitions (solid line). Pronounced steepening of the slope of $\delta(N)$ with increasing density indicates that Δ and ν'' have different signs but are similar with respect to absolute values. Let us now assume that in addition to collision broadening there exists some other cause of broadening of the nonlinear power resonance—for example, field broadening—which in the absence of collisions does not lead to line displacement. Inasmuch as the total width of resonance γ increases with increasing intensity, the foregoing reasoning clearly establishes that the region where the $\delta(N)$ curve inflects (Fig. 1) is shifted in the direction of smaller densities of the disturbing gas.

The new position of curve $\delta(N)$ is indicated by a broken line in Fig. 1. Thus, as the field intensity changes at a constant pressure of the perturbing gas, a transition from one curve to the other occurs which indicates increasing line displacement. Naturally, a similar effect of an increasing line displacement at a constant pressure occurs in the course of broadening caused by transit of a molecule through the optical beam or for some other reason. It should be emphasized that the effect is completely associated with the nonlinearity of the dependence of collision displacements on density N.

3. Leaving detailed consideration to subsequent publication, we shall adduce formulas and numerical calculations of the displacement of the case of relatively small densities when $\gamma/\Delta\omega_D\bar{\theta} \ll 1$.

We shall first consider the field broadening effect. Keeping first-order terms with respect to parameter $\gamma/\Delta\omega_D\bar{\theta}$, we shall obtain an expression for δ in the rate equation approximation for the displacement of the maximum of Lamb's dip in the absorption of a plane standing wave

$$\delta = \Delta + \nu'' \left[1 - \pi \frac{(\Gamma + \nu')\sqrt{1+2I}}{\Delta\omega_D\bar{\theta}} \; \frac{2 + 2\sqrt{1+2I}}{1 + 3\sqrt{1+2I}} \right], \tag{2}$$

where $I = (dE/\hbar)^2(\Gamma + \nu')^2$ is the saturation parameter, d is the transition dipole matrix element, E is the standing wave amplitude, and $\hbar$ is Planck's constant.

From all the lines which are used to stablize the laser frequency, the most completely understood so far is the $F_2^{(2)}$ component of the $P(7)$ vibrational-rotational line of the ν_3 band of methane ($\lambda = 3.39\ \mu$m). In the case of internal pressure broadening, $d\Delta/dN$ for this line is 100 Hz/mtor, $d(\Delta + \nu'')/dN = 10$ Hz/mtor,[1] and $\Gamma + \nu' = \Delta\omega_D\theta$ at $N \approx 10$ mtor.[6] This permits us to use Eq. (2) to calculate the variation in the magnitude of the line shift when the saturation parameter varies from zero to unity. At $N = 1$ mtor, $\delta(I = 1) - \delta(I = 0) = 15$ Hz; at $N = 3$ mtor, $\delta(I = 1) - \delta(I = 0) = 135$ Hz.

Allowance for transit broadening is made in the third-order theory of field perturbtions. The transverse field distribution of a standing wave was considered to be Gaussian. With accuracy up to first-order terms in parameter $\gamma/\Delta\omega_D\bar{\theta} \ll 1$, the shift of Lamb's dip maximum is

$$\delta = \Delta + \nu'' \left\{ 1 - \pi \frac{\Gamma + \nu'}{\Delta\omega_D\bar{\theta}} f\,[(\Gamma + \nu')\tau_0] \right\}, \qquad (3)$$

$\tau_0 = a/v_0$, a is Gaussian beam radius; Figure 2 shows the function $f(x)$. We shall calculate for the same methane line the variation in displacement as the parameter $(\Gamma + \nu')\tau_0$ varies from 1 to 2. At $N = 1$ mtor we get $\delta[(\Gamma + \nu')\tau_0 = 1] - \delta[(\Gamma + \nu')\tau_0 = 2] \approx 7$ Hz; at $N = 3$ mtor, $\delta[(\Gamma + \nu')\tau_0 = 1] - \delta[(\Gamma + \nu')\tau_0 = 2] \approx 60$ Hz.

The dependence of the position of the resonance maximum on such microscopic values as the saturation parameter and optical beam diameter is the basic factor which limits the frequency reproducibility of a laser standard. In the case involving the $F_2^{(2)}$ component of methane, the contribution of the effect considered here to such a dependence is less than the contribution caused by the presence of magnetic hyperfine line structure.[2,3] An attempt was made, in the hope of a substantial reduction in the displacement, to change to stabilization of the same transition in methane with respect to

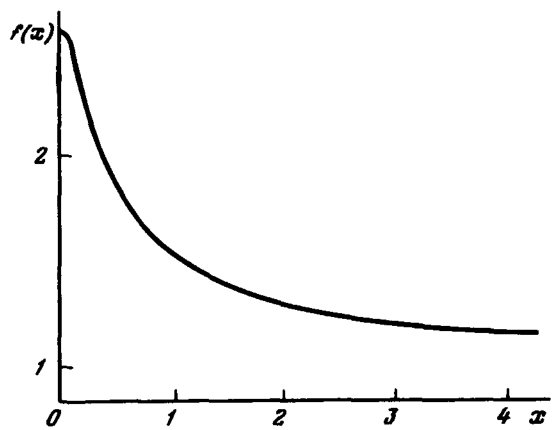

FIG. 2.

E for which the hyperfine structure is nonexistent.[8] The dependence of the line displacement on the microscopic parameters—considered in this paper—is, apparently, fundamental for this component as it is for the majority of other molecular transitions which are used for stabilization.

The authors thank I.I. Sobel'man for useful discussions.

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