Observation of narrow resonances in the output spectrum of a laser with nonlinear absorption

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The first direct observation of a new type of nonlinear-absorption resonance is reported. This is a resonance in the output spectrum of a laser with a nonlinear absorption which occurs when the width of the output line is significantly greater than the width of the resonance. Experiments show that at a low gas pressure the width of the resonance for vibrational-rotational transitions of molecules is determined exclusively by the collisional width.

1. The narrowing of saturated-absorption resonances in gases is accompanied by a sharp decrease in the intensity of these resonances because of decreases in the density of absorbing particles and in the intensity of the saturation field. This circumstance complicates the detection of resonances with relative widths less than 10^{-11}. Qualitatively new physical methods must accordingly be developed for producing resonances and detecting them if we wish to make further progress in ultrahigh-resolution spectroscopy. Another important problem in ultrahigh-resolution spectroscopy is to produce tunable light sources with a very narrow line (less than 100 Hz). In the present letter we offer the first report of the observation of a saturated-absorption resonance of a new type: a resonance in the output spectrum of a laser with an in-resonator absorbing medium. The width of this resonance corresponds to the homogeneous line width. In contrast with other methods of ultrahigh-resolution spectroscopy, which require continuously tunable single-frequency light with a narrow spectrum, this new method uses a laser whose output line width is substantially greater than the width of the resonance. This method has been used for the first experimental study of the behavior of the width of a resonance at low gas pressures. The results show that the width is determined exclusively by collisions.

2. The resonance in the output spectrum of a laser with a nonlinear absorption which was discussed in Ref. 1 results from a resonant dependence of the saturation absorption and of the refractive index in a gas in the field of a standing wave. Resonances of the absorption and of the refractive index are ordinarily observed in nonlinear ultrahigh-resolution spectroscopy.[2] If the width of the laser output line is much greater than the homogeneous width of the absorption line, it becomes impossible to observe the resonant dependence of the output power or frequency. However, resonances of the type described above arise in the output spectrum. Near the center of the absorption line the laser output spectrum is described by[1]

$$S(\Omega) \approx S_0\left[1+\frac{k}{1+(\Omega/\Gamma)^2}\right]\left[1+q\,\frac{1-(\Omega/\Gamma)^2}{\left(1+\left(\frac{\Omega}{\Gamma}\right)^2\right)^2}\right], \tag{1}$$

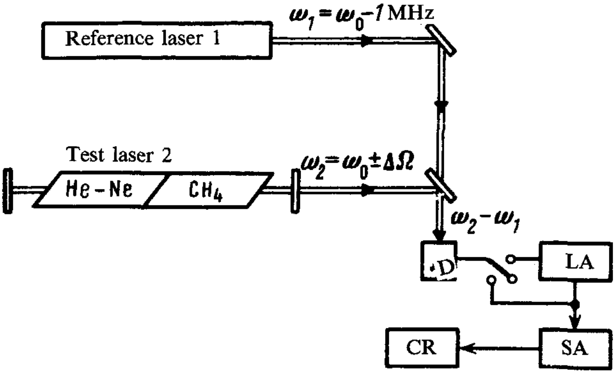

FIG. 1. Optical layout for observing the resonance in the output spectrum. D—Photodetector; LA—limiter amplifier; SA—spectrum analyzer; CR—chart recorder ($\Delta\Omega = 500$ kHz).

where S_0 is the spectral density of the laser output in the absence of absorption, $\Omega = \omega - \omega_0$ is the deviation of the output frequency ω from the center of the line (ω_0), and Γ is the homogeneous line half-width. The first and second terms correspond to resonances of the absorption and of the refractive index. The coefficients k and q determine the contrast of the power resonance and the coefficient of the nonlinear frequency pulling in a laser with nonlinear absorption.[2] At $\Gamma < 10^6$ Hz the output spectrum can be studied more conveniently by a heterodyne method (Fig. 1) with the help of an rf spectrum analyzer. If the width of the output line of a reference laser is much smaller than the homogeneous width 2Γ, the observed spectrum will correspond to the spectrum of the laser under study. Figure 2 shows the output spectrum of a He-Ne laser at $\lambda = 3.39$ μm with a methane absorber. The lengths of the resonator, of the amplifier tube, and of the absorption cell are 600, 200, and 400 cm, respectively. The

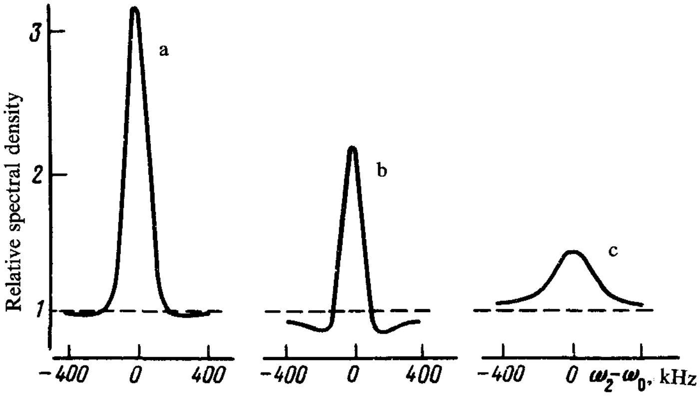

FIG. 2. Shapes of the resonance in the output spectrum of the He-Ne/CH_4 laser in various measurement regimes ($p_{CH_4} = 2$ mtorr). a—The beat output signal from the photodetector is fed directly to the spectrum analyzer; b—the beat signal is fed to the spectrum analyzer through a limiter amplifier; c—power resonance.

radius of the light beam in the cell is $a = 0.3$ cm. The width of the laser output line is ≈ 1 MHz. The resonance in the spectrum (Fig. 2) has a complex shape, which differs qualitatively from that described by (1). If the signal from the detector is fed to the spectrum analyzer through a limiter laser, the resonance observed in the laser output spectrum in this case will result exclusively from a nonlinear pulling of the output frequency toward the center of the absorption line [the second term in (1)]. The shape of the resonance in the output frequency spectrum is shown in Fig. 2b. The curve in Fig. 2c, found by dividing the curves in Figs. 2a and 2b, describes the effect of a change in the output power on the laser spectrum [the first term in (1)]. It can be seen from Fig. 2 that the changes in the power and in the refractive index make comparable contributions to the resonance in the output spectrum.

3. The high sensitivity of this method has made it possible to directly observe the magnetic hyperfine structure of the $F_2^{(2)}$ line of methane and a recoil effect and to study the behavior of the resonance at a low gas pressure, at which the transit time ($\tau = a/v_0$) of a particle moving at the average thermal velocity v_0 across the region of the interaction with the field (over a distance a) is significantly smaller than the reciprocal of the half-width Γ ($\Gamma\tau \ll 1$). While the width of the Lamb dip depends on the parameter Γ and on τ (Refs. 3–5), the width of the resonance in the spectrum is determined exclusively by the width 2Γ in the region $\Gamma\tau \ll 1$, as an analysis shows. A similar effect has been predicted for the width of the derivative of the saturation resonance with respect to the frequency[4] and for the resonance in the absorption coefficient.[6] An experimental study of this effect is clearly of interest, since it raises the possibility of producing narrow resonant lines with a width not restricted by transit broadening. Experimental difficulties have previously prevented anything beyond a qualitative observation of a "narrowing" of the resonance.[7,8]

Figure 3 shows the resonance half-width γ in the laser output spectrum versus the parameter $\Gamma\tau$. The solid line is calculated, while the points are experimental. The numerical calculation was carried out for a beam with a Gaussian profile; the hyperfine structure of the $F_2^{(2)}$ line of CH_4 was taken into account. The parameter $\Gamma\tau$ was

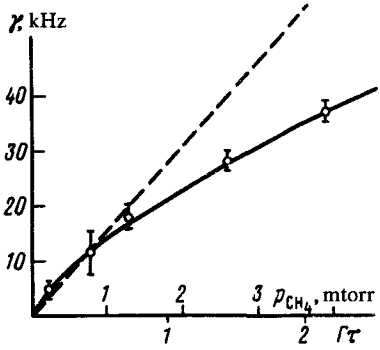

FIG. 3. Half-width of the resonance in the output spectrum versus the methane pressure (the parameter $\Gamma\tau$). Points—Experimental; solid line—theoretical; dashed line—collisional broadening of the resonance ($\gamma = \Gamma$). $a = 0.31$ cm, $\tau = 5.6 \times 10^{-6}$ s.

determined from the known collisional broadening in methane,[9] 15 kHz/mtorr, and the measured beam radius in the cell. The theoretical and experimental results agree well within the measurement errors. At values $\Gamma\tau \lesssim 1$ the width of the resonance is determined by the combined effects of collisional and transit-time broadening. At $\Gamma\tau \lesssim 0.1$ the half-width γ of the resonance in the output spectrum is directly related to the homogeneous half-width Γ ($\gamma \approx 1.4\ \Gamma$), and the transit-time effects are seen only in a pressure dependence of the slope of the broadening. The observed value of γ is 4 kHz at a methane pressure of 0.2 mtorr, or five times smaller than the transit-time half-width for particles with the average thermal velocity. Although the intensity of the resonance in the spectrum decreases in proportion to the square of the particle density ($\sim N^2$) in the transit region ($\Gamma\tau \ll 1$), the high sensitivity of this method made it possible to study the behavior of the homogeneous width of the resonance at low gas pressures.

In summary, these experiments demonstrate that this new ultrahigh-resolution method is highly effective. When combined with absolute measurements of the frequency in a heterodyne measurement method and with the use of tunable lasers, this resonance in the output spectrum may prove an effective method for ultrahigh-resolution spectroscopy for studying the saturated absorption and two-photon resonance in spatially separated optical fields.

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