Nonlinear Frequency-Dependent Diffraction Effect in Intracavity Resonance Asymmetries

A. Le Floch
Laboratoire d'Electronique Quantique, Université de Rennes, F-35042, Rennes, France

and

R. Le Naour
Laboratoire d'Electronique Quantique, Université de Nantes, F-44072, Nantes, France

and

J. M. Lenormand and J. P. Taché
Laboratoire d'Electronique Quantique, Université de Rennes, F-35042, Rennes, France
(Received 7 February 1980)

The asymmetry observed in intracavity resonances (especially of the Lamb-dip type) is attributed to a resonant nonlinear diffraction effect. Direct evidence of this effect is obtained by a new experiment and careful analysis of the diffracted light, in the geometrical shadow of the laser beam inside the cavity, which reveals a reversed asymmetry of the resonance. A modified Lamb-dip formula gives better agreement between experiment and theory. The effect occurs in many experiments with Gaussian beams, particularly in recent high-sensitivity intracavity laser spectroscopy.

PACS numbers: 42.60.He, 42.55.Bi

The asymmetry of atomic and molecular resonances is a ticklish problem which frequently occurs in laser physics. In particular, a careful analysis of the Lamb dip observed at the center of the Doppler profile of the output intensity of a monomode laser,[1-3] or of saturated absorption peaks,[4] often reveals indeed an evident asymmetry. The physical origin of this asymmetry has beem discussed extensively, experimental observations and theoretical interpretations remaining difficult. Such an asymmetry is important from a fundamental point of view and also in applications like metrology. This arises again in recent high-sensivity intracavity absorption spectroscopy.[5]

Many theoretical models explaining this asymmetry have been proposed such as a shift of the Lorentzian with respect to the center of the Doppler line by collision effects[6] or as a lens-type effect due to a radial atomic distribution in the laser medium. In this latter model where saturation is assumed unimportant, Casperson and Yariv[7] explain the typical higher intensity maximum on the low-frequency side than that on the high-frequency side by an enhancement of the mode volume, i.e., an enhancement of the number of interacting atoms due to a dispersion defocusing on this low-frequency side, associated with the atomic distribution. This mechanism, however, does not explain on one hand the asymmetry which remains with a homogeneous atomic distribution. On the other hand, it is in disagreement with the forward-diffracted-light analysis, which implies on the contrary a decrease of the mode volume on the low-frequency side. In this essentially nonlinear problem, taking account of the saturation by the Gaussian distribution of light and of the fundamental diffraction effect occurring in all resonators, we explain the output and diffracted beam asymmetries. Agreement between theory and experiment is then obtained.

We describe here an experiment (Fig. 1) which enables simultaneous comparison between the asymmetry of the laser intensity and that of the measured diffracted light inside a laser cavity. To avoid spurious effects the laser must be limited to monomode TEM_{00} operation. A 1.7-mm-diam aperture, matched to the mode size, enables TEM_{00} oscillation on the 3.39-μm line of a He^3-Ne^{20} laser at tube center, giving a controlled Gaussian shape intensity distribution in the beam section. The inner tube diameter ($\simeq$7 mm) is chosen greater than the mode section in order to provide a quasiconstant atomic distribution in this mode section centered on the tube axis giving an unsaturated gain such as $\Delta G_0/G_0 \lesssim 6\times10^{-2}$ and to avoid not-well-known edge effects. Detector D_1 measures the output intensity of the laser, while an identical D_2 detector measures the intensity of the diffracted light recovered by a thin golden mica sheet M_3, in the geometrical shadow of the aperture, without perturbing the laser itself. The output power is about 10 μW for a total pressure of 0.55 Torr in the tube, with a 7:1 mix-

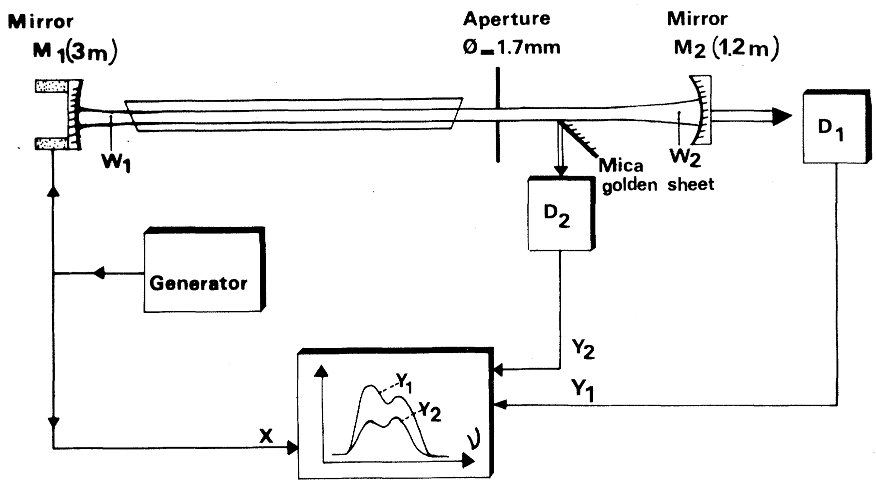

FIG. 1. Experimental setup.

ture. When the laser frequency is scanned, simultaneous recordings of both signals reveal a surprising result: The asymmetry of the diffracted light is quite different and is reversed as shown by the experimental curve of Fig. 2. Moreover, the Lamb-dip center of the output intensity is shifted towards the high-frequency side, while that of the diffracted beam is slightly shifted in opposite sense with respect to the center ν_0 of the laser line, defined as mid-distance between the threshold points. These results are inconsistent with the preceding interpretations. The collision model would lead to the same asymmetry on the two beams, and the Casperson-Yariv model would lead to an enhanced asymmetry of the same sign on the diffracted beam. On the contrary, the inverted observed asymmetries and the opposite shifts of the two different Lamb dips suggest that the physical origin of the asymmetry is to be assigned here to a frequency-dependent diffraction effect. This mechanism is explained and confirmed by further quantitative evaluations.

Variations of the normalized diffraction losses as a function of frequency are indeed easily obtained in our experiment by the diffracted inten-

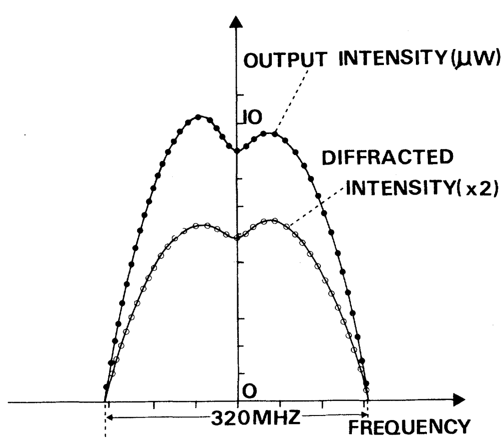

FIG. 2. Theoretical curves and experimental points of the output laser intensity and of the diffracted-light intensity. Both curves are fitted with B varying between extrema values 0.566 and 0.576, $C = 0.14$, and $D = 0.19$. The cavity length is 0.37 m and the mirror transmittance is 5%.

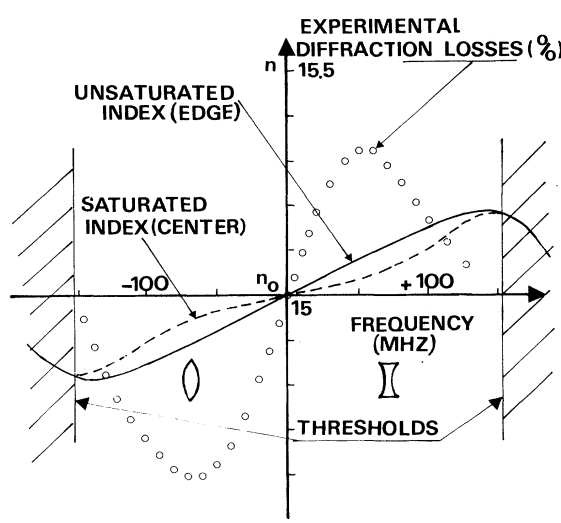

FIG. 3. Experimental variations of the normalized diffraction losses at the aperture vs frequency, with saturated and unsaturated dispersion curves, respectively, for the center and the edge of the beam section.

sity ratio, as shown in Fig. 3. The absolute values are obtained by comparison with calibrated losses like optical plates on one hand and by theoretical evaluation on the other hand.[8] The mechanism is explained in Fig. 3. The Gaussian distribution of the beam intensity saturates the dispersion curve more on the axis than on the edge of the laser mode.[9] It results that on the low-frequency side of the Lamb dip the saturated medium will focus the beam, while on the high-frequency side it will defocus it, leading, respectively, to lower or greater diffraction losses essentially located at the aperture in our case. The experiment effectively isolates the corresponding dispersion shape diffraction effect with relatively important signals and now provides us with a precise test to verify the agreement between theory and experiment for both experimental curves, when the measured effect is introduced in laser theory. We must then modify the usual frequency-dependent laser intensity formulas derived by different authors. In the case of linear polarization,[10] Lamb's self-consistent third-order perturbation calculation[11] which is valid at low power and low pressure, as in our experiment, led Szöke and Javan[12] to propose a formula modified by Cordover and Bonczyk[2], i.e.,

$$I_1(X-E)=A[1-B\exp(X^2)]/D_1(X-E), \qquad (1)$$

where the ratio B = losses/unsaturated gain is a constant, $X=(\nu-\nu_0)/\Delta\nu_D$, and

$$D_1(X-E)=1-\frac{2C}{\sqrt{\pi}}+D\frac{[1+(2C/\sqrt{\pi})]C^2}{C^2+(X-E)^2}.$$

The numerator in (1) is of Gaussian type while the denominator D_1 contains the Lorentzian responsible of the Lamb dip.[13] The phenomenological E parameter, introduced by collisional processes, shifts the Lorentzian from the center of the line and describes the Lamb-dip asymmetry. On the contrary, the diffraction effect measured in our experiment implies the introduction of frequency-dependent losses, that is $B \equiv B(X)$ is no longer constant and the preceding formula is to be written

$$I_2(X)=A[1-B(X)\exp(X^2)]/D_2(X), \qquad (2)$$

where $D_2(X)$ is now independent of E. From the measured variations of the diffraction losses we deduce the related $B(X)$ variations and it is then possible to trace the theoretical curves corresponding to the experiment. They are reported in Fig. 2 for the frequency dependence of the output laser intensity and of the diffracted intensity. The diffracted intensity is obtained by multiplying the $I_2(X)$ intensity by the measured aperture transfer function. Good agreement with experimental points for both curves is then observed. The asymmetries observed here are no longer due to collisional shift of the Lorentzian, but rather to an asymmetry of the Gaussian related to resonant diffraction losses which occur in the numerator of formula (2) of the output intensity. Moreover, the Lamb-dip shifts for both curves are effectively of opposite signs.

Considering with more precision the central region of the output intensity curve, one can ask if, for a given rate asymmetry defined as the difference between the heights of the maxima divided by their average height, the Lamb-dip shifts introduced by both formulas (1) and (2) are equal. In Fig. 4 we have expanded the central region of the experimental curve. It shows that for this relatively large asymmetry (7%) the two formulas lead to two distinguishable curves and that one with the diffraction effect gives clearly a better agreement with the experiment. The real shift due to the asymmetry is $\Delta_2 = 3.5 \pm 0.5$ MHz on the output intensity while the Lorentzian shift associated with the formula (1) would be $\Delta_1 = 12 \pm 0.5$ MHz for the same asymmetry. In addition to other possible shifts to ν_0 itself, this shift is particularly important in defining a frequency reference for a resonance which is not quite symmetric. Similar shifts observed in intracavity spectroscopy will prevent accurate wavelength measurements. The frequency-dependent diffraction effect may also introduce asymmetries and

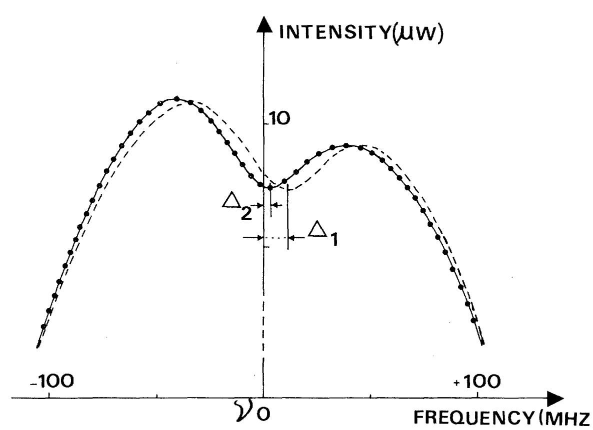

FIG. 4. Expanded Lamb-dip region for an observed $\Delta I/I \simeq 7\%$ asymmetry. Full line, theoretical curve given by formula (2); dashed line, theoretical curve given by formula (1); dotted line, experimental results.

frequency shifts in saturated absorption experiments especially when the gas cell is placed inside the laser cavity. Experiments performed with iodine, for example, seem to confirm this point.[4] In this case, however, the asymmetry on the output intensity is reversed, the dispersion curve being reversed for an absorption line.

The corresponding blue enhancement associated to absorption resonances appears again in recent experiments relative to the intracavity spectroscopy by laser quenching.[5] It limits the accuracy of this high-sensitivity spectroscopy (enhancement of 10^4 to 10^5) which allows in particular the detection of highly forbidden optical transitions. The diffraction experiment performed by ourselves with a Ne absorption cell placed inside a multimode dye laser oscillating around the 6143-Å line confirms the line shapes observed on the output beam by the preceding groups.[5] The blue enhancement and the sharp rise on the high-frequency side of the absorption line on the output beam correspond to a blue decrease and a sharp fall on the diffracted beam intensity. This suggests a new truthful method for intracavity absorption spectroscopy by combining the two signals. The original symmetry of the resonance is then restored, its shift vanishes and the possibilities of this spectroscopy are greatly improved. Symmetric Lamb-dip–type resonances can also be obtained by the same method from the curves of Fig. 2.

In conclusion we suggest a straightforward way to determine the effects of both gain saturation and intracavity diffraction. The direct analysis of the resonant diffracted light in the geometrical shadow of the laser beam leads to the origin of the asymmetry of intracavity resonances and particularly of the Lamb dip. The relatively large diffraction effect introduced in the theory gives better agreement with experiment than that obtained with earlier formulas. Diffracted-light analysis appears as a differential method and a powerful tool in studying and eliminating asymmetry problems which may occur in systems where "atoms–Gaussian beam–diffracting element" interactions exist.[14]

The authors are indebted to P. Brun for help in the work. This research was partially supported by the Centre National de la Recherche Scientifique.

[1] W. R. Bennett, Jr., S. F. Jacobs, J. T. La Tourette, and R. Rabinowitz, Appl. Phys. Lett. 5, 56 (1964); P. W. Smith, J. Appl. Phys. 37, 2089 (1966).

[2] R. H. Cordover and P. A. Bonczyck, Phys. Rev. 188, 696 (1969).

[3] C. Freed and H. A. Hauss, IEEE J. Quantum Electron. 9, 219 (1973).

[4] P. Cerez, A. Brillet, S. Hajdukovic, and N. Man, Opt. Commun. 21, 332 (1977).

[5] R. G. Bray, W. Henke, S. K. Liu, K. V. Reddy, and M. J. Berry, Chem. Phys. Lett. 47, 213 (1977); W. T. Hill, R. A. Abreu, T. W. Hänsch, and A. L. Schawlow, Opt. Commun. 32, 96 (1980); P. Kumar, G. O. Brink, S. Spence, and H. S. Lakkaraju, Opt. Commun. 32, 129 (1980).

[6] S. G. Rautian, Zh. Eksp. Teor. Fiz. 24, 788 (1967) [Sov. Phys. JETP 24, 788 (1967)]; P. R. Berman and W. E. Lamb, Jr., Phys. Rev. A 2, 2435 (1970), and 4, 319 (1971).

[7] L. W. Casperson and A. Yariv, Appl. Opt. 11, 462 (1972).

[8] A. E. Siegman, *An Introduction to Lasers and Masers* (McGraw-Hill, New York, 1971), p. 313; T. Li, Bell Syst. Tech. J. 44, 917 (1965).

[9] Such a spatially inhomogeneous saturation is supposed by C. Freed and H. A. Hauss (Ref. 3) but they do not take account of diffraction, and by B. K. Garside, IEEE J. Quantum Electron. 11, 940 (1968), for multimode spectra analysis.

[10] The analysis is restricted to the linearly polarized laser with its particular Lamb dip. See A. Le Floch, R. Le Naour, and G. Stephan, Phys. Rev. Lett. 39, 1611 (1977).

[11] W. E. Lamb, Jr., Phys. Rev. 134, 1429 (1964).

[12] A. Szöke and A. Javan, Phys. Rev. 145, 137 (1966).

[13] The Doppler-limit approximation is valid since $C = \gamma_{ab}'/kv_m \simeq 0.14$, in agreement with Sargent and Scully, in *Laser Handbook*, edited by F. T. Arrechi and E. O. Schulz-Dubois (North-Holland, Amsterdam, 1972), Vol. 1, p. 81.

[14] It might occur with progressive electromagnetic fields but we note its nonreciprocal character, the diffraction element being necessarily placed after the atomic or molecular system.