edited by A. Mooradian, T. Jaeger, and P. Stokseth (Springer-Verlag, Berlin, 1976), p. 122.

[3]A. Lauberau and W. Kaiser, in *Chemical and Biochemical Applications of Lasers*, edited by C. Bradley Moore (Academic, New York, 1977), Vol. 2, p. 87.

[4]D. A. Angelov *et al.*, Proceedings of the European Physical Society International Conference on Lasers in Photomedicine and Photobiology, Florence, 3–5 September 1979 (to be published).

[5]R. Rigler, in *Chromosome Identification*, Nobel Symposium 23, edited by T. Casperson and L. Zech (Academic, New York, 1973), p. 335.

[6]A. Andreoni, C. A. Sacchi, and O. Svelto, in *Chemical and Biochemical Applications of Lasers*, edited by C. Bradley Moore (Academic, New York, 1979), Vol. 4, p. 1.

[7]J. Piette, C. M. Calberg-Bach, and A. van de Vorst, Photochem. Photobiol. 26, 377 (1977).

[8]R. Cubeddu and S. De Silvestri, Opt. Quant. Electron. 11, 276 (1979).

[9]R. Cubeddu, S. De Silvestri, and O. Svelto, to be published.

Doppler-Free Two-Photon Dispersion and Optical Bistability in Rubidium Vapor

E. Giacobino, M. Devaud, F. Biraben, and G. Grynberg

Laboratoire d'Optique Quantique du Centre National de la Recherche Scientifique, Ecole Polytechnique, 91128 Palaiseau Cédex, France, and Laboratoire de Spectroscopie Hertzienne de l'Ecole Normale Supérieure, Université Pierre et Marie Curie, 75230 Paris Cédex 05, France

(Received 11 March 1980)

Doppler-free two-photon dispersion has been observed in rubidium vapor near the $5S_{1/2}-5D_{5/2}$ two-photon transition. The experiment was performed by looking at the shape of the transmission peaks of a Fabry-Perot cavity filled with rubidium. When the nonlinear susceptibility is large, a bistable behavior occurs. A nonperturbative treatment of the susceptibility is required for the interpretation of these results.

PACS numbers: 42.65.Gv, 32.80.Kf

When a nonlinear medium is placed inside a Fabry-Perot cavity, there can be several solutions for the transmitted beam power with the same value of incident power. In the case of two stable solutions, the phenomenon is known as optical bistability.[1,2] There has been a large amount of interest in this domain[3] since the first experiment of Gibbs, Mc Call, and Venkatesan.[2] In all the experiments performed in atomic vapors, the frequency of the beam was near a one-photon allowed transition of the atoms. In that case the nonlinearity arises from the saturation of the absorption or of the dispersion. A different possibility[4,5] has been theoretically discussed: It consists of the use of two-photon absorption[4] or two-photon dispersion.[5,6] In that case, even if the transition is not saturated, the response of the vapor is obviously nonlinear. Moreover, since bistability is generally observed in the standing wave of a linear cavity, the Doppler broadening of the two-photon transition is cancelled under this geometry.[7,8] Thus we can expect a very rapid change of the bistable behavior in a range of a few megahertz near the two-photon resonance. We report in this paper the results of an experiment performed in order to verify this effect.
As shown below, the experiment has brought several valuable pieces of information, both on Doppler-free two-photon dispersion and on optical bistability.

An interesting feature of the two-photon transition is that all the atoms interact in the same way with the field either in absorption or in dispersion.[5] This is due to the cancellation of the Doppler effect: All the atoms, whatever their velocities may be, have the same energy detuning from the two-photon resonance.[9,10] In comparison with experiments performed with a single-photon transition,[2] it results in two advantages[5]: (1) There is no average on the Maxwell velocity distribution. The atomic response is given by a simple analytic formula. (2) It is possible to reach high values of the nonlinear refractive index because the energy detuning can be of the order of the natural width.

The experiment can be performed with the usual setup for Doppler-free two-photon spectroscopy.[9,11] The light coming from a cw dye laser is sent into an experimental cell placed inside a Fabry-Perot cavity. In previous experiments, bistability was not observed because the nonlinear susceptibilities were too small. We have chosen to perform the experiment in rubidium around the $5S_{1/2}$-$5D_{5/2}$ two-photon transition (λ

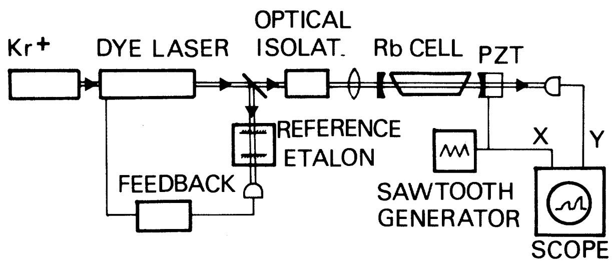

FIG. 1. Experimental setup. The single-mode dye laser is servo-locked on the transmission peak of an external reference etalon which can be pressure tuned.

$\simeq 7779$ Å). This is a very intense two-photon transition because the oscillator strengths f_{5S-5P} and f_{5P-5D} are large and because the energy detuning from the one-photon transition $5S_{1/2}-5P_{3/2}$ is small (~ 35 cm^{-1}). Furthermore, it is possible to obtain a large density of rubidium atoms at a relatively low temperature. It can be noticed that at 180 °C the two-photon absorption leads to a fluorescent line at 4201 Å ($6P_{3/2}\rightarrow 5S_{1/2}$), coming in a cascade from the $5D_{5/2}$ level, which is visible to the naked eye.

The experimental setup is shown in Fig. 1. We use a single-mode oxazine dye laser (built accordingly to the design of Pinard, Aminof, and Laloë[12]) pumped by a Kr^+ ion laser. We obtain about 150 mW at 7779 Å. The light is transmitted through a Faraday rotator in order to prevent the reflected beam from coming back into the laser cavity. The Rb cell is made of fused silica and the windows are tilted at Brewster's angle. The Fabry-Perot cavity is 15 cm long. The mirrors have 96% reflectivity and their radii of curvature are choosen in order to give a Rayleigh range of the order of 5 cm for the Gaussian fundamental mode. One of the mirrors is mounted on a piezoceramic transducer (PZT). The length of the cavity is continuously scanned by applying a symmetrical sawtooth on the PZT. A signal proportional to the transmitted light is obtained with a fast photodiode and is sent on the Y channel of an oscilloscope. A signal proportional to the voltage applied to the PZT is sent on the X channel. We observe two peaks on the oscilloscope: They correspond to the increase and to the decrease of the length of the cavity. They do not overlap because of the hysteresis of the PZT.

The shape of the transmission peaks can be theoretically predicted by use of the construction of Marburger and Falber[13] which is applied to our problem in Fig. 2. If the nonlinear suscepti-

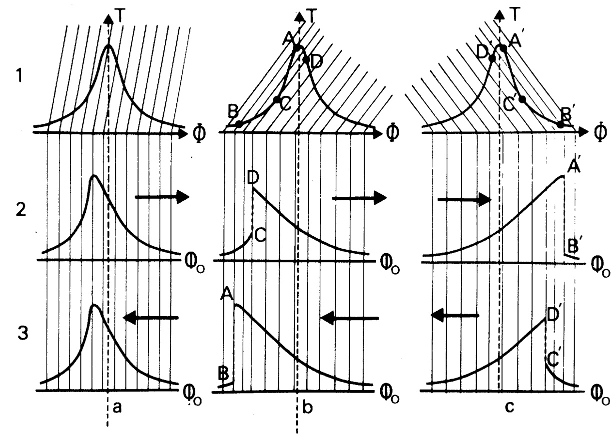

FIG. 2. Theoretical construction of the transmission of a Fabry-Perot cavity filled with a nonlinear medium. In the diagrams of line 1, we have first plotted the transmission T of the cavity against the phase Φ of the electric field after one round trip (Airy curve). But, because of the nonlinear medium, Φ itself depends on T. ($\Phi=\Phi_0+\chi_{\rm NL}'\omega l/c$, where l is the length of the cavity and Φ_0 is the round-trip phase in absence of nonlinear effects. To the lowest order, $\chi_{\rm NL}'$ is proportional to the intensity inside the Fabry-Perot cavity. Thus we obtain for Φ a straight line as a function of T. The law becomes $\Phi=\Phi_0+\eta T$, where η is proportional to the incident intensity and to $\chi_{\rm NL}'$.) The transmission corresponding to a given value of Φ_0 is given by the intersection of the Airy curve with the straight line. Varying the length of the cavity corresponds to varying Φ_0 while leaving the slope constant. From these remarks it is easy to construct the plot of the transmission as a function of Φ_0 or, what is the same thing, of the length of the cavity (lines 2 and 3). (a) When the slope of the straight line is large (low incident intensity and nonlinear index), there can be only one crossing point between the Airy curve and the straight line, whatever its position: The peaks only look asymmetrical. (b) When the slope is lower than that of the inflection tangent of the Airy curve, there can be three crossing points. The one situated on the portion of the Airy curve between points A and C is an unstable solution, while the others are stable. If the straight line is moved from the left to the right of the diagram, the transmission increases from B to C, then jumps to D and decreases. If the straight line is moved in the other direction, the transmission increases up to A, then jumps to B. Thus we obtain two different shapes for the two scanning directions. (c) A similar argument can be developed when the slope of the straight line has the opposite sign. The steep edges appear on the other side of the transmission peak.

bility $\chi_{\rm NL}'$ is zero, both peaks are symmetrical and identical to each other, and correspond to the usual shape of the Airy peaks. If $\chi_{\rm NL}'$ increases, the following behavior can be theoretically predicted: First the transmitted peaks become asymmetrical [Fig. 2(a)]. Then for higher values of χ_{NL}' we expect a different shape for the two peaks; they should have different heights and show a sharp edge which corresponds to the transition from one stable point to another [Figs. 2(b) and 2(c)].

We did observe this behavior experimentally (Fig. 3). We show in Fig. 3 three sets of oscilloscope traces of the transmission peaks, obtained at three different temperatures (165, 180, and 200 °C) for different energy detunings from the two-photon resonances (as there are two isotopes and many hyperfine components, our experiment corresponds to ^{85}Rb and to the transition starting from the $F=3$ hyperfine ground sublevel).

It can be seen that at low temperature and for large energy detunings, the transmission peaks look symmetrical. On the other hand, for small values of the energy detuning an obviously bistable behavior appears. One feature is very surprising: The sharp edge is always on the same side of the peak. It means that χ_{NL}' keeps the same sign in the whole range of frequencies where bistability is observed {if the sign of χ_{NL}' changes, the slope of the straight line in Fig. 2 changes which leads to a transmission peak with the sharp edge on the other side [Fig. 2(c)]}. More precisely, from the experimental data, we can determine that χ_{NL}' is positive.

Such a behavior cannot be understood if the nonlinear susceptibility is calculated in the framework of lowest-order perturbation theory. In that case, we obtain for χ_{NL}' a curve which has a dispersive shape: Its sign changes for the value of the frequency which corresponds to the two-photon resonance. However, the experimental effects can be accounted for by use of theories developed previously.[4,5]

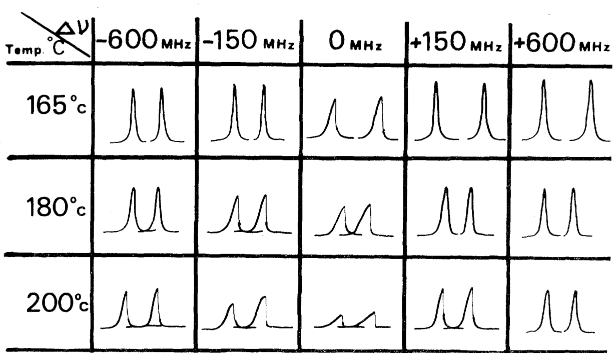

FIG. 3. Oscilloscope traces of the transmission when the length of the cavity is swept with a sawtooth. We have used various oven temperatures, 165, 180, and 200°C, corresponding to the rubidium densities (2, 4, and 8)$\times 10^{14}$ atoms/cm^3, respectively, and studied the bistability effect for various detunings $\Delta\nu$ ($\Delta\nu = -\delta E/h$) of the incident wavelength from the two-photon resonance. On each diagram the first trace from the left corresponds to an increasing length of the cavity, the second trace to a decreasing length.

We have calculated χ_{NL}' when the atomic system can be considered as a two-level system connected by a two-photon transition. Such a system can be exactly solved and we have found for χ_{NL}' the formula

$$\chi_{NL}' = \frac{N}{\epsilon_0}\frac{|Q_{ge}|^2 I(\delta E - 2s\Gamma_{eg}/\Gamma_e)}{\delta E^2 + \hbar^2\Gamma_{eg}^{\;2}(1+a^2)}.$$

In this formula N is the density of atoms; Q_{ge} is the matrix element of the two-photon operator[14] $Q = \vec{D}\cdot\vec{\epsilon}(\hbar\omega - H_0)^{-1}\vec{D}\cdot\vec{\epsilon}$, where $\vec{D}$ is the electric dipole operator, ϵ the polarization of the electric field, and H_0 the Hamiltonian of the free atom; I is the intensity of the electric field; $\delta E = \hbar\omega_{eg} - 2\hbar\omega + 2s$ is the energy detuning from the two-photon resonance with the light shift s of the transition, $s = \frac{1}{4}(Q_{ee}' - Q_{gg})I$, taken into account where

$$Q' = \vec{D}\cdot\vec{\epsilon}(E_e - \hbar\omega - H_0)^{-1}\vec{D}\cdot\vec{\epsilon} + \vec{D}\cdot\vec{\epsilon}(E_e + \hbar\omega - H_0)^{-1}\vec{D}\cdot\vec{\epsilon};$$

Γ_{eg} and Γ_e are the relaxation rates of the optical coherence and of the excited state, respectively; and a is the saturation parameter of the two-photon resonance,[15] whose value is $|Q_{eg}|I/\hbar(\Gamma_e\Gamma_{eg})^{1/2}$.

There are two terms in χ_{NL}'. The sign of the first one, proportional to δE, changes at resonance. It corresponds to the polarization of the medium induced by the two-photon transition. The second term, proportional to $-2s\Gamma_{eg}/\Gamma_e$, appears at a higher perturbation order (s is proportional to I). This term can be interpreted as a modification of the linear susceptibility due to the new distribution of population in the ground and excited levels in the presence of the quasiresonant two-photon excitation. It must be noticed that the sign of the second term does not change with $\omega_{eg} - 2\omega$.[16] Let us now estimate the relative magnitude of these two terms. The ratio of their maxima is of the order of $s/\hbar\Gamma_e(1+a^2)^{1/2}$. For the present experimental condition, a is larger than 1 and this ratio can be shown to be of the order of

$$(f_{5S_{1/2}-5P_{3/2}}/f_{5P_{3/2}-5D_{5/2}})^{1/2}(\Gamma_{eg}/\Gamma_e)^{1/2}.$$

Thus it is dependent on temperature because Γ_{eg}

depends on collisional effects and increases with the number of rubidium atoms. In our experimental conditions, the value of the ratio is of the order of 10. This means that the second term of χ_{NL}' is predominant. Even if the sign of χ_{NL}' changes for a large value of δE, the corresponding values of χ_{NL}' are too small to induce bistability. The theory thus permits one to understand why χ_{NL}' apparently always keeps the same sign. Moreover, the light shift can be theoretically estimated. Its value is negative and the factor $-2s\Gamma_{eg}/\Gamma_e$ is positive in agreement with the experimental observation.

On the other hand, the curves corresponding to opposite values of the energy detuning are different. This is because on the one side of the resonance δE and s have the same sign and their effects add up whereas on the other side of the resonance the two contributions subtract from one another.

In conclusion, we have observed Doppler-free two-photon dispersion for the first time. We have shown that it is important to describe the system with a nonperturbative method in order to understand the experimental observations. Lastly, we have shown that this effect can be applied to the observation of optical bistability.

We would like to thank Professor B. Cagnac, Dr. C. Flytzanis, Dr. J. P. Hermann, and Dr. P. W. Smith for the stimulating discussions concerning this experiment.

[1]Phys. Today 30, No. 12, 17 (1977).

[2]J. M. Gibbs, S. L. McCall, and T. N. C. Venkatesan, Phys. Rev. Lett. 36, 1135 (1976).

[3]P. W. Smith and E. H. Turner, Appl. Phys. Lett. 30, 280 (1977); T. N. C. Venkatesan and S. L. McCall, Appl. Phys. Lett. 30, 282 (1977); T. Bishofberger and Y. R. Shen, Appl. Phys. Lett. 32, 156 (1978).

[4]F. T. Arecchi and A. Politi, Lett. Nuovo Cimento 23, 65 (1978).

[5]G. Grynberg, M. Devaud, C. Flytzanis, and B. Cagnac, to be published.

[6]P. F. Liao and G. C. Bjorklund, Phys. Rev. Lett. 36, 584 (1976), and Phys. Rev. A 15, 2009 (1977).

[7]L. S. Vasilenko, V. P. Chebotayev, and A. V. Shishaev, Pis'ma Zh. Eksp. Teor. Fiz. 12, 161 (1970) [JETP Lett. 12, 113 (1970)]; B. Cagnac, G. Grynberg, and F. Biraben, J. Phys. (Paris) 34, 845 (1973).

[8]F. Biraben, B. Cagnac, and G. Grynberg, Phys. Rev. Lett. 32, 643 (1974); M. D. Levenson and N. Bloembergen, Phys. Rev. Lett. 32, 645 (1974).

[9]G. Grynberg and B. Cagnac, Rep. Prog. Phys. 40, 791 (1977).

[10]M. Bassini, F. Biraben, B. Cagnac, and G. Grynberg, Opt. Commun. 21, 263 (1977).

[11]E. Giacobino, F. Biraben, G. Grynberg, and B. Cagnac, J. Phys. (Paris) 38, 623 (1977).

[12]M. Pinard, C. G. Aminof, and F. Laloë, Appl. Phys. 15, 371 (1978).

[13]J. H. Marburger and F. S. Felber, Phys. Rev. A 17, 335 (1978).

[14]G. Grynberg, F. Biraben, E. Giacobino, and B. Cagnac, J. Phys. (Paris) 38, 629 (1977).

[15]G. Grynberg, J. Phys. (Paris) 40, 657 (1979).

[16]In addition, the construction we give in Fig. 2 must be slightly modified: As the light shift s depends on the intensity, the law giving Φ versus the transmission (due to the nonlinearity of the medium) cannot rigorously be represented by a straight line (see Ref. 5). Nevertheless in our experimental conditions, this effect has no significant influence on the shape of the transmission peaks of the cavity.