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P. Pignolet, B. Dubreuil, P. Davy, J. Minard. Observation of splitting and optical satellites on the $H\delta$ line of hydrogen due to non-resonant interaction with a CO_2 laser radiation. *Journal de Physique Lettres*, 1981, 42 (10), pp.205-209. 10.1051/jphyslet:019810042010020500 . jpa-00231910

HAL Id: jpa-00231910

<https://hal.science/jpa-00231910>

Submitted on 1 Jan 1981

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Classification
Physics Abstracts
32.70 — 32.90

Observation of splitting and optical satellites on the H_δ line of hydrogen due to non-resonant interaction with a CO_2 laser radiation (*)

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(Reçu le 2 février 1981, accepté le 27 mars 1981)

Résumé. — On a étudié expérimentalement l'effet Stark dynamique non résonnant des états $n_a = 6$ et 5 de l'hydrogène quand les états excités d'une décharge luminescente d'hydrogène sont perturbés par le faisceau d'un laser T.E.A. à CO_2 . En particulier, les déplacements des niveaux $n_a = 6$ et 5 (respectivement $0,21 \text{ cm}^{-1}/\text{MW}/\text{cm}^2$ et $-0,08 \text{ cm}^{-1}/\text{MW}/\text{cm}^2$) sont mesurés et les effets de la levée de la dégénérescence et de l'émission de composantes satellites sont observés sur l'aile bleue de la raie H_δ . Un traitement perturbatif de l'interaction permet de rendre compte des effets observés.

Abstract. — The non-resonant A.C. Stark effects of the $n_a = 6$ and 5 manifolds of hydrogen have been studied when excited atoms of a hydrogen glow discharge are irradiated by a T.E.A. CO_2 laser beam. Shifts, respectively $0.21 \text{ cm}^{-1}/\text{MW}/\text{cm}^2$ for $n_a = 6$, $-0.08 \text{ cm}^{-1}/\text{MW}/\text{cm}^2$ for $n_a = 5$ are measured, and splitting and optical satellites originating from high orbital quantum numbers are observed on the blue wing of the H_δ line. Restructuring of this line is described by a perturbative treatment of the interaction.

In an early experiment, we succeeded in measuring the shift of the H_δ line of hydrogen due to non-resonant perturbation of the $n_a = 6$ manifold by a c.w. CO_2 laser radiation [1], the so-called light-shift or non-resonant A.C. Stark effect. With laser energy flux I_L in the $1\text{--}5 \text{ MW}/\text{cm}^2$ range, the measured shifts ($\Delta E_6 \lesssim 1.1 \text{ cm}^{-1}$) were in qualitative agreement with theoretical calculations of the A.C. Stark effect developed within the framework of second order perturbation theory [2, 3]. Later on, B. A. Zon [4, 5] reconsidered these calculations taking into account the degeneracy in the orbital moment l_a , which was neglected in the earlier studies. In particular, B. A. Zon [4] pointed out that the non-resonant interaction of a laser radiation with an hydrogen state $|n_a, l_a, m_a\rangle$ (m_a : magnetic quantum number) leads not only to coupling with other states $|n_b, l_b, m_b\rangle$, ($b \neq a$) but also to mixing of the orbital states $|n_a, l'_a, m'_a\rangle$ ($l'_a \neq l_a$) corresponding to the same principal quantum number n_a . In this case, the quasi-stationary states for laser fields with polarization vector $\mathbf{e}$ are constructed

from an expansion of the atomic wave functions (atomic units are used) :

$$\psi_{n_a}(t) = \sum_{l_a} a_{l_a m_a}(t) e^{-iE_{n_a}t} |n_a, l_a, m_a\rangle + \sum_{\substack{n_b, l_b, m_b \\ \neq n_a, l_a}} b_{n_b l_b m_b}(t) e^{-iE_{n_b}t} |n_b, l_b, m_b\rangle \quad (1)$$

$\psi_{n_a}(t)$ is a solution of the time-dependent Schrödinger equation including the interaction operator

$$\tilde{V}(t) = R_e(\mathbf{d} \cdot \mathbf{F}_L e^{-i\omega_L t})$$

where $\mathbf{d}$ is the atomic dipole moment, $\mathbf{F}_L$ and ω_L are respectively the amplitude and frequency of the laser electric field. A perturbation calculation, valid for $\tilde{V}(t) \ll E_{n_a+1} - E_{n_a}$ and for spontaneous and ionization level widths smaller than the detuning from resonance, then leads to the solution :

$$\psi_{n_a}(t) = \sum_k C_k e^{-i(E_{n_a} + \Delta E_{n_a}^{(k)})t} \sum_{l_a m_a} g_{l_a m_a}^{(k)} |n_a, l_a, m_a\rangle \quad (2)$$

where k , $g_{l_a m_a}^{(k)}$ are respectively the number and the eigen-vectors of the matrix $[\phi]$:

$$\phi_{l_a m_a l'_a m'_a} = \sum_{n_b l_b m_b} \frac{\langle n_a, l_a, m_a | \mathbf{d} \mathbf{e} | n_b, l_b, m_b \rangle \langle n_b, l_b, m_b | \mathbf{d}^+ \mathbf{e}^* | n_a, l'_a, m'_a \rangle}{E_b - E_a + \hbar\omega_L} + \frac{\langle n_a, l_a, m_a | \mathbf{d}^+ \mathbf{e}^* | n_b, l_b, m_b \rangle \langle n_b, l_b, m_b | \mathbf{d} \mathbf{e} | n_a, l'_a, m'_a \rangle}{E_b - E_a - \hbar\omega_L}$$

and C_k are integration constants.

(*) Work partly supported by D.R.E.T. under contract n° 79/151.

The shift $\Delta E_{n_a}^{(k)}$ of quasi-stationary states

$$\varphi_{n_a}^{(k)} = \sum_{l_a m_a} g_{l_a m_a}^{(k)} |n_a, l_a, m_a\rangle$$

is related to the eigen-value $\beta_{n_a}^{(k)}$ of $[\phi]$ through the relation :

$$\Delta E_{n_a}^{(k)} = -\frac{F^2}{4} \beta_{n_a}^{(k)}.$$

For the case of a linearly polarized field, $\mathbf{e} = \mathbf{z}$ is the quantization axis, whereas for a right or left-handed circularly polarized field, $\mathbf{e} = \frac{\mathbf{x} \pm i\mathbf{y}}{\sqrt{2}}$ the quantization axis is clearly defined in the direction of propagation of the radiation. The magnetic sublevels are not then mixed by the field and quantum numbers n_a, m_a well characterize the quasi-stationary states. The following selection rules hold :

$$m_a = m'_a = m_b$$

for linear polarization and $m_a = m'_a = m_b \pm 1$ for right or left-handed polarization.

If the field is elliptically polarized

$$\mathbf{e} = \frac{\mathbf{x} + i\mathbf{y}}{\sqrt{2}} \cos \theta + \frac{\mathbf{x} - i\mathbf{y}}{\sqrt{2}} \sin \theta$$

(where $\cos \theta$ determines the degree of circular polarization of the radiation), there is no axial symmetry (states with $|m_a - m'_a| = 2, 4, \dots$ are mixed by the field) and there are no selection rules in m_b .

Non-resonant interaction with the laser field then leads to a splitting of the hydrogen spectrum which was predicted in the early calculations [2, 3]. Furthermore, a consequence of the formation of new states $\varphi_{n_a}^{(k)}$ in the laser field is the removal of the orbital momentum selection rules. In particular, for the $n_a = 6 \rightarrow n'_a = 2$ transition leading to emission of the H_δ line, B. A. Zon [5] showed that optical satellites starting from F, G and H sublevels of $n_a = 6$ were sufficiently separated from the allowed line and strong enough to be observed with a linearly polarized CO_2 laser flux $I_L > 1 \text{ MW/cm}^2$. In the general case of a partially polarized field, states with different m_a are also strongly mixed [6] so that the field completely removes the degeneracy in m_a . For these quasi-stationary states no selection rules hold and they all can decay into a given channel giving rise to additional satellites. Energy of these quasi-stationary states indeed depends on the degree of polarization and varies between the two limiting cases of linearly and circularly polarized fields. Satellites, as well as splitting in the case of level shifts of the same order of magnitude as the Doppler width at room temperature ($\approx 0.7 \text{ cm}^{-1}$), are expected to give rise to broadening and distortion of the blue wing of the shifted H_δ line as deduced from the perturbed energy diagram shown in figure 1 and computed from references [2,

Fig. 1. — Perturbed energy diagram of H_δ line computed for $I_L = 1 \text{ MW/cm}^2$ for the linearly polarized CO_2 laser field. m is the magnetic quantum number and e (even), o (odd) indicate the parity of the sublevels.

3, 5], for $I_L = 1 \text{ MW/cm}^2$ for the linearly polarized CO_2 laser radiation ($\nu_L = 944 \text{ cm}^{-1}$)⁽¹⁾. Such an appearance was in fact observed in reference [1], but the large broadening was attributed to the strong spatial inhomogeneity of the laser beam intensity and to focusing conditions.

Similar calculations made for the $n_a = 5$ manifold indicate that the H_γ line (transition $n_a = 5 \rightarrow n'_a = 2$) does not show such a broadening but only a mean shift of the whole profile ($\overline{\Delta E}_5 \approx -0.07 \text{ cm}^{-1}$ when $I_L = 1 \text{ MW/cm}^2$).

In this paper, we describe a new experiment in which we study the shifts and the restructuring of the H_δ and H_γ ($\lambda_\delta = 4101.7 \text{ \AA}$) ($\lambda_\gamma = 4340.4 \text{ \AA}$) lines of the hydrogen atom in nearly uniform laser irradiation conditions and in the $I_L < 10 \text{ MW/cm}^2$ range. Comparison between these two different lines allows us to control the experimental conditions and to demonstrate without any doubt the splitting and satellite effects on the laser perturbed H_δ line.

The experiment is based on measurements of the spectrum of the light emitted by a glow discharge in hydrogen during interaction with a CO_2 T.E.A. laser beam. In a recent study, we used a low repetition rate (0.05 Hz) T.E.A. CO_2 laser constructed in our laboratory and producing 2 J in a 100 ns main pulse to study the non-resonant A.C. Stark effect of the most intense

⁽¹⁾ Note that the $n' = 2$ level A.C. Stark effect is small enough to be neglected compared to the spin-orbit coupling. This level acts as a detection level of the $n_a = 6$ perturbation.

lines of hydrogen (H_α , H_β) and helium ($\lambda = 5876 \text{ \AA}$, $5016 \text{ \AA}$) [7]. The GEN-TEC DD 250 T.E.A. CO_2 laser available in the present experiment produces 600 mJ in a 150 ns main pulse at $\lambda = 10.59 \text{ \mu m}$. Working at a repetition rate of 10 Hz, with shot-to-shot variation of the laser power within 10 % and jitter restricted to 10 ns, accumulation techniques have been developed to

enhance S/N ratio and detectivity of weak lines. The experimental set-up is shown in figure 2.

Excited states are populated in a capillary glow discharge (inner diameter 4 mm, length 20 mm). The discharge is created in high purity hydrogen gas by current pulses (intensity : 1-5 A, width = 80 μs) superimposed on a weak D.C. current ($i \approx 10 \text{ mA}$).

Fig. 2. — Experimental set-up : 1) discharge power supply ; 2) T.E.A. CO_2 laser ; 3) Monochromator ; 4) Monitoring system and gated integrators ; 5) Recorder ; 6) I.R. Monochromator ; 7) P.M. power supply.

Pressure can be adjusted from 0.1 to 10 torr. Characteristics of the ionized gas have been determined elsewhere [8] : electronic density $n_e < 5 \times 10^{12} \text{ cm}^{-3}$, mean electronic energy $\approx 3\text{-}8 \text{ eV}$, gas temperature $\approx 340 \text{ K}$.

The partially polarized CO_2 laser beam is partially focused along the discharge tube in order that the whole volume inside the capillary tube is irradiated. In the present experiment, the mean laser intensity I_L in the central part of the tube is limited to $6 \pm 1 \text{ MW/cm}^2$ and is measured at the output of the discharge tube taking into account transmission of the NaCl window. Observation of the laser intensity at the output of the discharge tube indicates a near uniform distribution in the central part (diameter $\approx 2 \text{ mm}$) viewed by the spectrometer.

The fluorescence light emitted by the discharge is observed axially and is imaged onto the slit of a 1.15 m S.O.P.R.A. motor-driven grating spectrometer and then onto a R.C.A. 7265 photomultiplier tube. Slit widths are adjusted in order to give a resolution of $0.12 \text{ \AA}$ in the vicinity of $\lambda = 4101.7 \text{ \AA}$. The line shapes are scanned at speeds as low as $0.08 \text{ \AA/min}$.

The whole system is synchronized by a clock which lights the discharge at 20 Hz and triggers the laser at 10 Hz. A two channel (A and B) gated integrator synchronized by this clock analyses the photomulti-

plier signal. The number of shots accumulated by the two integrators, as well as processing of the output signal are controlled by a M6800 microprocessor evaluation board. Gate A, 100 ns wide, is temporally positioned about the peak of the laser pulse, the discharge being on. Alternately, gate B, similar to A, is opened only when discharge is on and laser off. Thus, for a given wavelength λ and a predetermined number of shots, one gets alternately at a rate of 10 Hz, the intensity $J(\lambda)$ of the laser perturbed line in A and the intensity $J'(\lambda)$ of the same but unperturbed line in B. The outputs of gates A and B are displayed on a two channel strip chart recorder. The perturbed and unperturbed line profiles are obtained simultaneously by slowly rotating the grating of the spectrometer. Each point on the profile corresponds to a summation over 100 or 200 shots, that is an accumulation time of 10 (20) s and a scanned spectral range of 0.013 (0.026) $\text{\AA}$ which is small enough to ensure no resolution loss.

Observations of the laser pulse shape indicate that during the 100 ns duration of a single measurement, $I_L(t)$ varies typically between 75 and 100 % of its peak value. This leads to a corresponding variation of the A.C. Stark shift which finally appears as an extra source of broadening of the observed line. When typical lifetimes of the shifted levels are lower than the

laser pulse variation, the observed fluorescence spectrum follows adiabatically the laser pulse. In this case, the net effect can be taken into account by averaging over the laser pulse intensity. In the opposite case, time correlations must be taken into account [9].

The adiabatic approximation holds for the studied levels since previous measurements of relaxation rates on atomic excited states in a similar hydrogen discharge indicate typical lifetimes of 10 ns when

$$P = 1 \text{ torr} \quad [8].$$

Figure 3 shows a typical experimental recording obtained on the H_δ line for

$$I_L (\text{mean value}) = 4.0 \pm 0.7 \text{ MW/cm}^2.$$

Fig. 3. — Experimental recording of the H_δ line for

$$I_L = 4 \pm 0.7 \text{ MW/cm}^2.$$

Also shown are the theoretical positions of the components of this line : (—) allowed components; (---) satellites, computed for a CO_2 laser field with 25 % linear and 75 % circular polarization.

Similar results have been observed for laser intensities up to $8.0 \pm 1.5 \text{ MW/cm}^2$. In figure 3, the peak of the laser perturbed line is shifted towards the blue wavelengths : $\overline{\Delta E}_6 \cong 0.83 \text{ cm}^{-1}$. Furthermore, a strong broadening and structure are observed on the blue wing which extends over $\cong 1 \text{ Å}$.

On this experimental recording, we have drawn the position of the allowed and satellite components of the perturbed line computed for a laser field with 25 % linear and 75 % circular polarization. These satellites originating from the shifted (l_a, m_a) sublevels form the major contribution to the blue wing of the perturbed line.

In the same experimental conditions, we also measured the perturbation of the H_γ line. Figure 4 shows a typical result obtained for

$$I_L = 4.0 \pm 0.7 \text{ MW/cm}^2.$$

The measured shift $\overline{\Delta E}_5 = -0.32 \text{ cm}^{-1}$ is in broad agreement with theoretical calculations [2, 3, 5] as reported in table I.

Fig. 4. — Experimental recording of the H_γ line for

$$I_L = 4 \pm 0.7 \text{ MW/cm}^2.$$

Same comments as for figure 3.

Table I. — Theoretical ($\overline{\Delta E}_{n_a}^{\text{TH}}$) and experimental ($\overline{\Delta E}_{n_a}^{\text{EX}}$) values of H_γ and H_δ hydrogen line shifts induced by T.E.A. CO_2 laser radiation for $I_L = 4.6$ and 8 MW/cm^2 .

$H_\gamma (n = 5 \rightarrow n' = 2)$		$H_\delta (n = 6 \rightarrow n' = 2)$		I_L (MW/cm^2)
$\overline{\Delta E}_5^{\text{TH}} (\text{cm}^{-1})$ (Ref. [2])	$\overline{\Delta E}_5^{\text{EX}} (\text{cm}^{-1})$	$\overline{\Delta E}_6^{\text{TH}} (\text{cm}^{-1})$ (Ref. [2])	$\overline{\Delta E}_6^{\text{EX}} (\text{cm}^{-1})$	
- 0.28	- 0.32	0.88	0.83	4 ± 0.7
- 0.42	- 0.48	1.32	1.19	6 ± 1
- 0.56	- 0.59	1.76	1.81	8 ± 1.5

In particular, as expected, no broadening of the red wing is observed on this line since for the $n_a = 5$ manifold, the relative splitting is lower than for $n_a = 6$ and allowed satellite components still remain grouped within the Doppler width.

Comparison of the non-resonant A.C. Stark effect for the H_γ and H_δ lines, whose splitting and restructuring are quite different, allows us to check in an independent way the experimental conditions of the interaction. Apparition of satellites and their influence on the $n_a = 6 \rightarrow n' = 2$ emission profile are then clearly demonstrated in agreement with theoretical calculations.

Acknowledgments. — The technical assistance of M. Dudemaine and J. C. Pellicer is gratefully acknowledged.

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