Polarization Spectroscopy of Atomic Oxygen by Dye and Semiconductor Diode Lasers.

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Summary. — We used polarization spectroscopy to perform a high-resolution investigation of atomic oxygen in the visible and near-infrared spectral region by means of dye and semiconductor diode lasers. We measured the fine structure of the highly excited $4d^5D$ level and resolved the hyperfine structure of the metastable $3s^5S_2$ level of ^{17}O . Isotope shift values are also reported for several transitions of the three stable oxygen isotopes (^{16}O , ^{17}O , ^{18}O). Polarization lineshapes are discussed and compared with those obtained in saturation techniques.

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1. – Introduction.

The spectroscopy of the oxygen atom has revealed new interesting aspects after the demonstration of the possibility to resolve sub-Doppler features of its spectrum [1, 2].

Different techniques have been applied to the high-resolution investigation of this atom. Several groups used a two-photon scheme to excite the atoms from the $2p^3P$ ground state to the $3p^3P$ state in gaseous discharges and in flames [3]. In that case, however, the resolution was limited by the linewidth of the pulsed lasers.

Champeau *et al.* [4] used Fabry-Perot spectroscopy to investigate the fine structure of excited levels in a discharge cooled by liquid nitrogen.

Continuous-wave sub-Doppler spectroscopy was performed first by means of intermodulated optogalvanic technique on visible transitions starting from highly excited states [1]. This allowed us to measure the fine structure [5] and isotope shifts [6] of the investigated transitions with an improvement of more than one order of magni-

tude with respect to conventional spectroscopy data [7]. Recently, the first resolution of the hyperfine structure of ^{17}O was reported for the excited metastable $^5\text{S}_2$ level [8]. This result was achieved by saturation spectroscopy using an AlGaAs semiconductor diode laser.

A limitation to the resolution achievable by saturation spectroscopy schemes is set by the presence of a background due to collisions which change the velocity of atoms excited by the pump beam. These collisions rethermalize the velocity distribution altered by the laser radiation and give rise to a saturation signal also when the laser frequency is out of resonance [9]. However, experimental conditions can be found which lead to a reduction of this pedestal by means, for example, of fast amplitude modulation of the pump beam [10] or high-current operation of the discharge [11].

In the present paper, we report the results of measurements performed on visible and near-infrared transitions of oxygen by means of sub-Doppler polarization spectroscopy (PS) [12]. The theory underlying this spectroscopic technique is well known in the literature [13-15] and is briefly summarized in the next section. As far as the velocity-changing-collisions (VCC) effects are concerned, the relevant point is that, since the atomic orientation, which is probed in polarization spectroscopy, is easily destroyed in collisions, the amplitude of the collisional background results to be greatly reduced.

Furthermore, with respect to techniques based on optogalvanic detection [16] such as intermodulated optogalvanic spectroscopy (IMOGS), PS is less sensitive to the regime of operation of the discharge; thanks to the high detection sensitivity, this allows to work at lower gas pressures, with a consequent reduction of the homogeneous broadening of the spectral lines.

Finally, a simplification of the spectra can be obtained by means of PS, as will be shown below, and complex spectra can be more easily analysed.

This effort to improve the resolution is justified by the size of the effects we want to observe. The fine-structure separations and the isotope shift are much smaller than the Doppler width. In particular, in ref. [2] it was suggested that the measured isotope shifts in oxygen could indicate a nuclear volume effect, which, in general, would be absolutely negligible in such a light atom. As is well known, the ^{16}O nucleus plays a central role in nuclear physics because of its «doubly magic» structure, with both proton and neutron closed shells. The accurate measurement of the optical isotope shifts, together with the data obtained in ref. [8] from the analysis of the hyperfine structure, can give information on this nucleus, complementary to the ones obtained by nuclear physics techniques.

2. - Experimental apparatus.

In this work, two different laser sources were used: for the investigations starting from $3p$ states (fig. 1), tunable radiation was provided by an Ar^+ ion pumped c.w. ring dye laser (Coherent 699-21). Using Rhodamine 6G and DMC dyes we could cover the $(580 \div 680)$ nm range. A spectral width of about 1 MHz was obtained by locking the laser frequency to an external reference cavity. To investigate the transitions in the near-infrared (around 777 nm) a commercial AlGaAs diode laser (Sharp LTO24) was used, operating at 780 nm at room temperature. In order to improve the spectral purity, the laser was mounted in an extended cavity configuration. The light from the

Fig. 1. – Relevant atomic levels of oxygen. The arrows denote the transitions in (nm) investigated using polarization spectroscopy.

diode laser was collimated onto a diffraction grating and the first-order diffracted beam was coupled back into the diode laser cavity itself, which had a reduced-reflection coating on the output facet. This configuration allowed to select a single mode of the laser and to reduce the laser linewidth to less than 1 MHz. Coarse tuning was achieved by changing the temperature of the diode along with the grating angle. Frequency scans of $\sim 10 \text{ GHz}$ were achieved by synchronously changing the length of the cavity with a piezoelectric transducer mounted on the grating, and the injection current of the laser. An intracavity beam splitter ($R \sim 30\%$) acted as output coupler. Two beams with a power of about 6 mW and 4.5 mW were available for the experiment. In comparison with dye lasers, the intensity stability of diode lasers is orders of magnitude higher. The consequent increasing of the signal-to-noise ratio allows to improve considerably the sensitivity also in nonzero background detection schemes such as in saturation spectroscopy. Furthermore, frequency modulation of these lasers allows to perform first derivative of lineshapes which result of interest in several applications as will be discussed later.

The calibration of the frequency scan for both lasers was provided by high-finesse Fabry-Perot interferometers with a free spectral range of 75 MHz. Wavelength calibration was obtained using a travelling Michelson interferometer with an accuracy of 1 part in 10^6 .

In polarization experiments, the laser beam was split into two beams of different intensities (see fig. 2): the stronger one (pump beam) was circularly polarized using a mechanically stressed quartz cube while the weaker (probe beam) was linearly polarized. The beams were sent in opposite directions through the sample cell. Changes of polarization of the probe beam were detected by means of a crossed polarizer with

Fig. 2. – Schematic experimental set-up for polarization spectroscopy (BS: beam-splitter, M: mirror, P: polarizer, Pt: phototube).

high extinction coefficient ($\sim 10^{-7}$) and the transmitted radiation collected on a photomultiplier tube. Residual birefringence of the optical windows of the sample cell was compensated by slightly squeezing the windows. Polarization signals could be detected directly on an oscilloscope or chart recorder. To increase the sensitivity, the pump beam was mechanically chopped and the probe signal analysed by means of a lock-in amplifier.

The sample cell was a 5 mm diameter pyrex tube. Atomic oxygen was produced by means of an argon-sustained electrodeless discharge where molecular oxygen was introduced at trace level. Details on the collisional physics leading to the formation of atomic oxygen are reported in ref. [17].

The discharge was normally operated at 1 Torr of buffer gas and 0.1 Torr of oxygen. For isotope shift investigations, ^{17}O - ^{18}O enriched samples were necessary, due to the low natural abundance of these isotopes ($^{17}\text{O} = 0.04\%$, $^{18}\text{O} = 0.2\%$).

3. – Theory.

Polarization spectroscopy is based on light-induced birefringence and dichroism of an absorbing gas. Such effects were observed for the first time by Gozzini [18] using classical radiation sources. Polarization spectroscopy with single-mode tunable lasers provides a very sensitive Doppler-free technique [12] allowing the investigation of extremely weak transitions.

As mentioned in the previous section, a typical experimental arrangement used in PS consists of a circularly polarized pump beam and a linear counterpropagating probe beam. In order to derive an expression for the polarization signal, in the following we assume the pump beam to be circularly polarized. The linearly polarized probe beam can be considered as the superposition of right (σ^+) and left (σ^-) circular polarizations. In the absence of the pump beam and when the laser frequency is resonant with the transition under investigation, the intensity transmitted by the crossed polarizer is

$$(1) \quad I_{(\text{pump off})} = I_0(\xi + \theta^2 + b^2) \exp[-\alpha L],$$

where I_0 is the probe beam intensity, ξ is the finite extinction coefficient, θ

is the angular deviation from complete crossing, and b is a term which takes into account the birefringence introduced by the windows of the sample cell.

When the laser frequency is tuned inside the homogeneous width of the line, σ^+ and σ^- components experience different absorption coefficients (α^+ and α^-) and refractive index (n^+ and n^-) due to the velocity-selective pumping caused by the σ^+ pump beam. As a consequence, after crossing the sample, the resulting probe polarization will be generally elliptical with the minor axis rotated with respect to the transmission axis of the analyser (x -axis in fig. 2). The component of the probe electric field E_0 transmitted along the x -direction is

$$(2) \quad E_x = E_0 \exp[i(\omega_0 t - kz)] \exp[i\{\omega_0/c(n^+ - n^-) + i(\alpha^+ - \alpha^-)/2\}L + b\}],$$

where L is the optical length of the sample and ω_0 is the line frequency.

In the limit of small saturation and thin sample, it can be shown that the intensity I of the polarization signal can be written as [14]

$$(3) \quad I = I_0 \exp[-\alpha L] \{ \xi + \theta^2 + b^2 - (1/2)\theta L \Delta\alpha [x/(1+x^2)] - \\ - (1/2)bL \Delta\alpha [1/(1+x^2)] + (1/16)(\Delta\alpha L)^2 [1/(1+x^2)] \},$$

where $x = (\omega - \omega_0)/\gamma$ is the laser detuning $\omega - \omega_0$ relative to the homogeneous width γ of the transition and $\Delta\alpha$ is the difference of the absorption coefficients for the σ^+ and σ^- probe components. In eq. (3) only $\Delta\alpha$ appears because Δn is related to $\Delta\alpha$ by the dispersion relations (Kramers-Kronig relations).

Equation (3) provides the spectral profile of the polarization signal; it is given by a constant term $I_0(\xi + \theta^2 + b^2)$, a dispersion shape term and Lorentzian shape terms. By an appropriate choice of the experimental conditions a pure Lorentzian or dispersive signal can be obtained. For example, if the polarizer is slightly rotated ($\theta \neq 0$) the dispersive shape may be made dominant. On the contrary, if the polarizers are completely crossed ($\theta = 0$), the lineshape is Lorentzian. Furthermore, if the pump intensity is sufficiently low ($b \Delta\alpha L \gg (\Delta\alpha L)^2$), the sign of the polarization signals depends on the difference

$$(4) \quad -\Delta\alpha = -(\alpha^+ - \alpha^-).$$

The calculation of the relative intensities in polarization spectra requires to estimate the difference in absorption for the two σ^+ and σ^- probe components. If we denote the pump and probe transitions as $J - J_1$ and $J - J_2$ respectively, then

$$(5) \quad \Delta\alpha = \sum_M n_M (\sigma_{JJ_2M}^+ - \sigma_{JJ_2M}^-),$$

where the sum is extended to the $(2J + 1)$ degenerate sublevels M in the lower pump level J . $\sigma_{JJ_2M}^\pm$ are the absorption cross-sections: their M dependences for $\Delta J = 1$ (P lines), $\Delta J = -1$ (R lines) and $\Delta J = 0$ (Q lines) are expressed in terms of Clebsch-Gordan coefficients. The population in the M -th sublevel is depleted by the circularly polarized pump wave according to

$$(6) \quad n_M = n_0 / (2J + 1)(1 + S_M),$$

where n_0 is the total density in the lower level and S_M is the saturation parameter for the M sublevel. The final result can be obtained making the sum over the Clebsch-Gor-

dan coefficients in eqs. (5) and (6):

$$(7) \quad \Delta\alpha = -\alpha_0 \mu_{JJ_1J_2} (I_1/S),$$

α_0 being the unsaturated probe absorption coefficient and I_1 the pump intensity. The expression for the saturation parameters S and the tabulated values for the anisotropy factor $\mu_{JJ_1J_2}$ are reported in ref.[13]. For $\Delta J = 0$ transitions, the anisotropy factor $\mu_{JJ_1J_2}$ results to depend on the quantum number J as $1/J^2$. To investigate $\Delta J = 0$ lines, it is more convenient to use a different scheme in which pump and probe beams are both linearly polarized with their polarization planes forming a $\pi/4$ angle. In this case, it can be shown that an expression similar to eq. (3) is obtained with the Lorentzian and dispersion terms interchanged.

It is worth observing that in the derivation of eq. (7) we have supposed that the probe transition is not coincident with the pump one. In the three-level scheme other kinds of resonances must be taken into account as, for example, optical-optical double resonances and crossover signals[19]. The former require two different laser frequencies (two-colour experiments) while crossovers can be observed halfway between transitions whose Doppler broadened profiles partially overlap.

4. - Results and discussion.

4.1. *Lineshapes study and comparison with saturation techniques.* - As discussed in sect. 3, polarization signals can be described as the superposition of a dispersive

Fig. 3. - Lorentzian (a) and dispersive (b) polarization signal for the 5S_2 - 5P_3 oxygen transition; c) is the derivative of the dispersive shaped profile.

and a Lorentzian shape. In fig. 3 we show typical lineshapes of the 5S_2 - 5P_3 oxygen transition at $\lambda = 777.4$ nm obtained in polarization spectroscopy by means of a diode laser. The two extreme regimes of symmetrical Lorentzian (fig. 3a)) and dispersive shapes (fig. 3b)) were achieved setting two different uncrossing angles θ between the two polarizers. Narrower lineshapes, down to one half of the natural width, can be obtained recording the first derivative of the dispersion-shaped signal, as shown in fig. 3c). The derivative of a dispersive line profile is given by

$$(8) \quad (d/dx)[x/(1+x^2)] = [1/(1+x^2)]^2 - [x/(1+x^2)]^2$$

and, hence, is a symmetric line centred at the same frequency of the line under investigation. The small asymmetry observed in fig. 3c) can be ascribed to the fact that the lineshape of fig. 3b) is not a pure dispersive profile. The «artificial» subnatural resolution achieved in this way can provide some advantages for high-precision frequency measurements or for laser locking. In addition, this technique allows to remove sloped background superimposed to Lorentzian lineshape with consequent improvements in resolution.

Fig. 4. – IMOGS (a)) and PS (b)) recordings for the $3p\ ^5P_3$ - $5s\ ^5S_2$ transition at 645.6 nm. The experimental lineshapes (triangles) are fitted with the superposition of Lorentzian and Gaussian curves in a) and a pure Lorentzian in b). (O_2 -Ar mixtures in the following ratios were used: $P_{O_2} = 0.1$ Torr, $P_{Ar} = 5$ Torr in a); $P_{O_2} = 0.05$ Torr, $P_{Ar} = 1.0$ Torr in b)).

It is well known in saturated spectroscopy that velocity-changing collisions can rethermalize the velocity distribution altered by the laser radiation. These collisions can modify the observed lineshape and are more or less effective depending on whether the detected observable is conserved or destroyed after the collision. In saturation spectroscopy the atomic populations are probed and VCC can represent a serious limit to resolution. Indeed, in these cases VCC make possible the simultaneous interaction of the probe and pump beam with atoms moving along the laser beams directions. As a consequence, a signal is detected also when the laser frequency is tuned out of the homogeneous resonance but still inside the Doppler profile. It has been shown that in the conditions of «strong» collisions [9] and assuming the homogeneous width γ much narrower than the Doppler $\Delta\nu_D$ width, the line profile is described by a Lorentzian peak superimposed over a Doppler pedestal. Typical recordings obtained using IMOGS and PS are shown in fig. 4 for the oxygen transition $3p\ ^5P_3-5s\ ^5S_2$ at 645.6 nm; the IMOGS lineshape is well fitted by the superposition of a Lorentzian and a Gaussian, as predicted by the theory. On the contrary, in the polarization spectrum (fig. 4b)) the absence of any Doppler background is an evidence of the fact that atomic orientation is destroyed during collisions. This feature is of obvious interest because it can simplify the spectroscopic resolution of closely spaced spectral components, as will be shown later.

4.2. Fine-structure measurements. – In a previous paper [5] the first study concerning the fine structure of the $^5D_{4,0}$ term has been reported using IMOG spectroscopy. In that work the fine-structure intervals were determined through the study of the $3p\ ^5P_{1,2,3}-4d\ ^5D_{4,0}$ multiplet at 616 nm and the values obtained were one order of magnitude more accurate than those obtained by Doppler-limited spectroscopy. However, the presence of the intense VCC background in IMOGS signal limited the resolution and could introduce slight shifts of the resonance centre frequencies due to the sloped background originated by adjacent fine-structure components. In order to improve accuracy and sensitivity, in this work we have remeasured such fine-structure separations using polarization spectroscopy. Figure 5 shows the recordings concerning the $^5P_1-^5D_{2,1,0}$, and $^5P_2-^5D_{3,2,1}$, and $^5P_3-^5D_{4,3,2}$ transitions.

The Doppler broadening for the typical temperature of our discharge ($T \sim 520$ K) is $\Delta\nu_D \sim 2$ GHz, which results comparable to the fine-structure intervals. A consequence of this is the appearance of crossover peaks in all the recordings in fig. 5. Differently from saturation spectroscopy, where real transitions and crossovers have the same phase, in polarization spectra positive and negative peaks can be observed. This feature is particularly useful to discriminate real lines and crossovers as explained in the following.

We limit our discussion to the $^5P_1-^5D_2$ transition and to the crossover between this transition and the $^5P_1-^5D_1$; an extension to other transitions is straightforward. Our analysis is similar to that reported in ref. [20] and will be described with the help of fig. 6. In order to give a phenomenological description of the different signs originated in PS spectra we remind that, under appropriate conditions, the sign of polarization signal is given by $-\Delta\alpha = -(\alpha^+ - \alpha^-)$. The σ^+ pump beam depletes the M sublevel by an amount proportional to the Clebsch-Gordan coefficients (values reported in brackets in fig. 6). The probe σ^+ component is less absorbed than the σ^- one because the M sublevels shared with the pump are less populated; since $\alpha^+ - \alpha^- < 0$, a positive polarization signal is generated.

The case of crossover peaks is depicted in fig. 6b). The pump and the probe beams,

Fig. 5. - Polarization spectra of the three fine-structure components arising from the ${}^5P_{1,2,3}$ levels (CO denote crossover peaks). In the lower part the oscillator strengths (continuous lines) are compared with the observed polarization intensities (dashed lines).

because of the Doppler shift, can interact simultaneously with atoms with velocity $v = c(\omega_{12} - \omega_{\text{laser}})/\omega_{\text{laser}} = c(\omega_{\text{laser}} - \omega_{11})/\omega_{\text{laser}}$. If the pump and probe waves are resonant, respectively, with 1-2 and 1-1 transitions, we obtain $\alpha^+ > \alpha^-$ and the crossover signal is then negative. This result is the same if pump and probe waves are interchanged in the scheme of fig. 6b). Of course, the intensity of the crossover signals depends on the number of atoms which have the right velocity to interact with both the beams; an overlapping function must then be considered, given by $\exp[-(|\omega_{11} - \omega_{12}|/2\Delta\omega_D)^2]$.

In the lower part of the recordings of fig. 5 the oscillator strengths and the experimental polarization intensities are compared. These diagrams show that polarization

Fig. 6. – Schematic illustration of the pumping effects determining the intensities of direct resonances (a)) and crossover signals (b)) in polarization spectroscopy.

intensities for $\Delta J = 0$ components are lower than the relative oscillator strengths and the difference increases with J . In the case of $\Delta J = \pm 1$ lines we found a good agreement between polarization signals and oscillator strengths. This is due to the similar M -dependence of the absorption cross-sections $\sigma_{JJ_2M}^+$ and $\sigma_{JJ_2M}^-$ for $\Delta J = 0$ transitions. In this case, and especially for high-angular-momentum states, where $\sigma_{JJ_2M}^+$ approaches $\sigma_{JJ_2M}^-$, it is more convenient to use the scheme of linearly polarized pump beam.

The data obtained for the fine-structure splittings 0-1, 1-2, 2-3, and 3-3 are reported in table I. Although the quoted standard deviations in this work and in ref. [5] are comparable, we attribute the discrepancies of the two sets of measurements to possible line shifts, as already discussed.

TABLE I. – *Fine-structure splittings for the $4d^5D$ oxygen level. The errors correspond to one standard deviation.*

Δ_{i-j}	This work (MHz)	Ref. [5] (MHz)
$\Delta_{(0-1)}$	787 (20)	735 (20)
$\Delta_{(1-2)}$	1422 (14)	1360 (20)
$\Delta_{(2-3)}$	1627 (20)	1594 (20)
$\Delta_{(3-4)}$	1347 (20)	1382 (20)

Accurate measurements of fine structure in atomic oxygen are of interest because oxygen is relatively light and, hence, theoretically still tractable. The theory of fine-structure in oxygen has been given in ref. [4], taking into account spin-orbit, spin-spin, and spin-other-orbit interactions. In that work it was suggested that fine-structure intervals can be described in terms of an analogous formula valid for the hyperfine interaction (Casimir formula):

$$(9) \quad W(\text{SLJ}) = (1/2)A'X + (3/8)B'[X(X+1) - (4/3)S(S+1)L(L+1)],$$

with $X = J(J+1) - L(L+1) - S(S+1)$. We have found that the fine-structure intervals of the quintet 5D term obey the Casimir formula. The estimated fine-structure constants A' and B' obtained by fitting the values of table I are $A' = -455(10)$ MHz and $B' = 31(2)$ MHz.

4'3. Isotope shift and hyperfine structure. – The «selection rules» valid in polarization spectroscopy offer the advantage of a drastic simplification in complex spectra, allowing easier assignments. In the specific case of isotope shift (IS) measurements, this turned out to be particularly important because it avoided overlapping of fine-structure components of different isotopes. In fig. 7, a comparison between IMOGS and PS spectra for the 5P_3 - 5D_J transition is shown, obtained using enriched ${}^{18}\text{O}$ samples. Due to the presence of fine-structure components and to the large Doppler pedestal, the resolution achieved in IMOGS is not high enough to resolve isotopic structure. In polarization spectroscopy only one of the three fine-structure components, namely the 5P_3 - 5D_4 , is clearly observed (see fig. 5c)) allowing an accurate measurement of the isotope shift.

In table II, we report the ${}^{16}\text{O}$ - ${}^{17}\text{O}$ and ${}^{16}\text{O}$ - ${}^{18}\text{O}$ IS values measured in this work for the $3s\ {}^5S_2$ - $3p\ {}^5P_1$ and $3p\ {}^3P_2$ - $5d\ {}^3D_{1,2,3}$ transitions. For completeness, we also report the IS values for other lines we measured by polarization spectroscopy and already re-

Fig. 7. – Comparison between IMOGS and PS recordings for the $3p\ {}^5P$ - $4d\ {}^5D$ transition ($\lambda = 615.8$ nm) obtained in ${}^{18}\text{O}$ enriched samples.

TABLE II. – *Isotope shifts for various transitions of atomic oxygen investigated by polarization spectroscopy. Quoted errors correspond to one standard deviation.*

Transition	Wavelength (nm)	IS (^{16}O - ^{17}O) (MHz)	IS (^{16}O - ^{18}O) (MHz)
$3p\ ^5P_2$ - $5d\ D_J$	595.9	746 (50)	1360 (50) this work
$3s\ ^5S_2$ - $3p\ ^5P_1$	777.7	1018 (45)	1970 (25) this work
$3p\ ^3P_2$ - $6s\ ^3S_1$	604.6	723 (10)	1278 (10) ref. [2]
$3p\ ^5P_3$ - $4d\ ^5D_4$	615.8	686 (10)	1320 (10) ref. [2]
$3p\ ^5P_3$ - $5s\ ^5S_2$	645.6	646 (10)	1163 (10) ref. [2]

ported in a previous paper [2]. Data on isotope shifts in oxygen are of great interest due to the importance of ^{16}O in nuclear physics. Recently, we have reported an experimental evidence of a field contribution to the isotope shift [2] which should be absolutely negligible for such a light atom. The analysis was performed following a parametric description based on King plots [21]. Confirmation of this effect has stimulated our group to extend this investigation to other levels particularly penetrating in the nucleus.

As is known, unlike even oxygen isotopes, ^{17}O has a nuclear spin $I = 5/2$. The only study of hyperfine structure of oxygen levels was performed on the 3P ground-state using nuclear magnetic-resonance techniques [22]. Recently, the first measurement of the h.f. structure of the excited 5S_2 level using saturated-absorption spectroscopy has been reported [8]. For all the transitions starting from $3p\ ^3P$ and $3p\ ^5P$ levels the hyperfine structure was not resolved because the homogeneous width (~ 150 MHz) was comparable with h.f. separations. The metastable 5S_2 level is of extreme interest in the oxygen spectra. It is the lowest S level in energy and, hence, it is more penetrating in the nucleus. As a consequence,

Fig. 8. – Polarization spectrum for the $3s\ ^5S_2$ - $3p\ ^5P_1$ transition ($\lambda = 777.7$ nm) recorded using an isotopically enriched sample (labelling numbers refer to the quantum number F of the lower 5S_2 level).

nuclear effects, such as field shift and hyperfine coupling, are expected to be particularly large.

In fig. 8 we show the polarization spectra relative to the 5S_2 - 5P_1 transition, recorded using the first derivative technique in an isotopically ^{17}O - ^{18}O enriched sample. Besides the resonance of the even isotopes ^{16}O and ^{18}O , the spectrum shows peaks due to the hyperfine structure of the odd ^{17}O isotope; it consists of five groups of peaks each of them corresponding to the hyperfine splitting of the lower 5S_2 level ($F = 1/2, 3/2, \dots, 9/2$). However, each of the peaks masks the hyperfine structure of the upper 5P_1 level. The value obtained from fig. 8 for the magnetic-coupling constant A for the 5S_2 level is $A = 74(15)$ MHz. This value results in agreement with that estimated in ref. [8].

5. - Conclusions.

Polarization spectroscopy has been applied to the study of atomic oxygen transitions in the visible and near-infrared spectral region. Accurate values for the fine-structure separations of the 5D level have been obtained. A comparison with a theoretical model has allowed to estimate the fine-structure constants.

Using appropriate polarization schemes it has been possible to introduce drastic simplification in the spectra. This allowed to measure isotope shifts for oxygen multiplets with fine-structure intervals comparable with isotope shifts.

The magnetic hyperfine-structure constant of the 5S_2 level has been remeasured; the values are in agreement with our recent estimate obtained using saturation spectroscopy. The accuracy of both the determinations are limited by the unresolved hyperfine structure of the upper level of the studied 5S_2 - 5P_J transition. Further investigations are in progress, in which different spectroscopic techniques are employed to improve the sensitivity and the spectral resolution [23].

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REFERENCES

- [1] M. INGUSCIO, P. MINUTOLO, A. SASSO and G. M. TINO: *Phys. Rev. A*, **36**, 4056 (1988).
- [2] L. GIANFRANI, A. SASSO, G. M. TINO and M. INGUSCIO: *Opt. Commun.*, **78**, 158 (1990).
- [3] D. J. BAMFORD, M. J. DYER and W. K. BISCHER: *Phys. Rev. A*, **36**, 3497 (1987); S. KROLL, H. LUNDBERG, A. PERSSON and S. SVANBERG: *Phys. Rev. Lett.*, **55**, 284 (1985); M. ALDEN, U. WESTBLOM and J. E. M. GOLDSMITH: *Opt. Lett.*, **14**, 305 (1989).
- [4] R. J. CHAMPEAU, J. JOCOTTON and E. LUC-KOENIG: *J. Phys. B*, **8**, 728 (1975).
- [5] A. SASSO, P. MINUTOLO, M. I. SCHISANO, G. M. TINO and M. INGUSCIO: *J. Opt. Soc. Am. B*, **5**, 1481 (1988).
- [6] K. ERNST, A. SASSO, G. M. TINO and M. INGUSCIO: *Opt. Lett.*, **11**, 554 (1989).

- [7] K. B. S. ERIKSSON and H. B. S. ISBERG: *Ark. Fys.*, **24**, 549 (1963); C. E. MOORE: *Natl. Stand. Ref. Data Ser. Natl. Bur. Stand.*, **35**, Vol. I, II, III.
- [8] G. M. TINO, L. HOLLBERG, A. SASSO, M. INGUSCIO and M. BARSANTI: *Phys. Rev. Lett.*, **64**, 2999 (1990).
- [9] P. W. SMITH and T. HANSCH: *Phys. Rev. Lett.*, **26**, 740 (1971).
- [10] P. HANNAFORD and D. S. GOUGH: in *EGAS Conference, Gratz 1988*.
- [11] A. SASSO, G. M. TINO, M. INGUSCIO, N. BEVERINI and M. FRANCESCONI: *Nuovo Cimento D*, **10**, 941 (1988).
- [12] C. WIEMAN and T. W. HANSCH: *Phys. Rev. Lett.*, **36**, 1170 (1976).
- [13] R. E. TEETS, F. V. KOWALSKI, W. T. HILL, N. CARLSON and T. W. HANSCH: *Proc. Soc. Photo-Opt. Instrum. Eng.*, **113**, 80 (1977).
- [14] W. DENTRODER: *Laser Spectroscopy* (Springer-Verlag, Berlin, Heidelberg, New York, N.Y., 1981).
- [15] N. A. CHEREPKOV and V. V. KUZNETSOV: *J. Phys. B*, **22**, L-405 (1989).
- [16] B. BARBIERI, N. BEVERINI and A. SASSO: *Rev. Mod. Phys.*, **62**, 603 (1990).
- [17] A. SASSO, M. I. SCHISANO, G. M. TINO and M. INGUSCIO: *J. Chem. Phys.*, **93**, 7774 (1990).
- [18] G. FORNACA, A. GOZZINI and F. STRUMIA: in *Electronic Magnetic Resonance and Solid Dielectric*, edited by R. SERVANT and A. CHURRU (North-Holland Publ. Comp., Amsterdam, 1963), p. 554.
- [19] S. NAKAYAMA: *Opt. Commun.*, **50**, 19 (1984).
- [20] CH. HERTZLER and H. J. FOTH: *Chem. Phys. Lett.*, **166**, 551 (1990).
- [21] J. BAUCHE and R.-J. CHAMPEAU: *Adv. At. Mol. Phys.*, **12**, 39 (1976).
- [22] J. S. M. HARVEY: *Proc. R. Soc. London, Ser. A*, **285**, 581 (1965).
- [23] M. DE ANGELIS, M. INGUSCIO, L. JULIEN, F. MARIN, A. SASSO and G. M. TINO: to be published.