NON LINEAR HANLE EFFECT : OPTOGALVANIC DETECTION AND APPLICATION TO POWER ENHANCEMENT IN GAS LASERS

F. Strumia

NON LINEAR HANLE EFFECT : OPTOGALVANIC DETECTION AND APPLICATION TO POWER ENHANCEMENT IN GAS LASERS

F. Strumia

Dipartimento di Fisica, P.za Torricelli 2, 56100 Pisa, Italy

Résumé - L'effet Hanle nonlinéaire est étudié à l'aide de la spectroscopie optogalvanique. L'accroissement prévu de la puissance émise des lasers à gaz est comparé aux données expérimentales.

Abstract - The Nonlinear Hanle effect was investigated by means of the optogalvanic spectroscopy. The predicted output power enhancement for gaseous lasers was compared with the available experimental results.

I - Non Linear Hanle effect

The Hanle effect was discovered sixty years ago [1] and consists in a change in the polarization of the resonance radiation when an external magnetic field removes the degeneracy between the Zeeman sublevels. With the advent of the laser the Non Linear Hanle Effect (NLHE) was also discovered [2]. It manifests as an increase of the saturated absorption when the M sublevels degeneracy is removed by means of a Zeeman or Stark effect.

As it is well known, in presence of a saturating laser beam of intensity I, the absorption coefficient of a Doppler broadened line can be written as

$$k(\nu,S) = \frac{k_o \, /\mu/^2}{\sqrt{1+S}} \quad \exp - \left(\frac{\nu - \nu_o}{\Delta \nu_D}\right)^2 \qquad 1)$$

where $k_o \, /\mu/^2$ is the absorption coefficient in absence of saturation and $S = I/I_S$ is the saturation parameter. Explicit expressions for I_S can be found in standard texbooks [3] and depend upon the relaxation processes in the system under investigation.

In the simplest case of a $J=0 \leftrightarrow 1$ transition and a laser beam linearly polarized orthogonal to the static field or propagating along the static field ($\Delta M=+1$ transitions) the absorption increase as a consequence of the NLHE can be easily calculated to be [4,5,6]

$$R(S) = \frac{k^F(S)}{k(S)} = \sqrt{\frac{1+2S}{1+S}} \qquad 2)$$

where $k^F(S)$ is the saturated absorption in presence of a static field large enough to resolve the M sublevels with respect to the homogeneous linewidth $\Delta \nu_H$, and $R(S)$ is the saturated absorption in absence of an external field. In eq. 2 it has been assumed a Doppler width much

larger than the homogeneous linewidth and a light beam of costant cross
section intensity. When the laser beam is polarized for inducing only
$\Delta M=0$ transitions $R^F(S) = R(S)$ and no absorption increase occurs as it
has been experimentally demonstrated /7/. The absorption increase as
expected from eq.2 is large and independent of the frequency setting
of the laser beam within the Doppler profile of the transition (fig.1a).
On the contrary a signal as shown in fig.1b is expected when the laser
frequency is set near the line center and the absorption signal is ob-
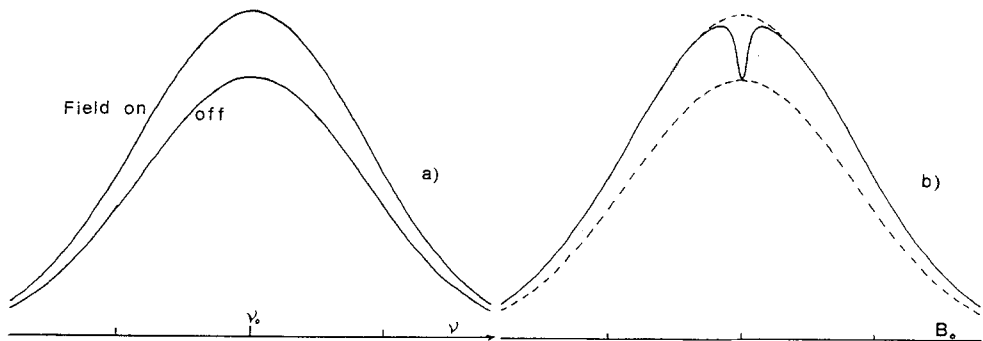served as a function of the applied external field. It consists of a

Fig. 1

difference between a Gaussian line due to the Zeeman scanning of the
Doppler line by the $\Delta M=\pm 1$ transitions and a Lorentzian central dip
which describes the NLHE.The HWHM of the Lorentzian is equal to the
FWHM of the J=1 level and is given by

$$HWHM = 2g_J\mu_0 B \qquad\qquad 3)$$

where g_J is the Landé factor and μ_0 the Bohr's magneton. In the more
general case of a $J \leftrightarrow J'$ transition, a density matrix treatment is ne-
cessary for describing the NLHE in both the upper and lower states of
the transition /4/, however for high J transitions such a treatment is
unpractical and cumbersome.

A simple calculation of the absorption increase in the high field li-
miting case (M degeneracy remove) is more convenient for understanding
the magnitude of the effect. In the absence of an external field the
natural quantization axis of the system is determined by the polariza-
tion and propagation direction of the laser beam. For a linearly polari-
zed beam and a dipole interaction the quantization axis is parallel to
EM field and only transitions with $\Delta M=0$ are induced. As a consequence
the saturation absorption coefficient is given by

$$R(J,S) = \sum_{-J}^{+J} R^0_M(S) = \sum_{-J}^{+J} \frac{|\mu^0_M|^2}{\sqrt{1 + \sigma|\mu^0_M|^2}} \qquad\qquad 4)$$

where the $|\mu^0_M|^2$ are the dipole moments for $\Delta M=0$ transitions computed by
means of the Clebsh-Gordan coefficients. This selection rule is con-

served if a static external field is applied parallel to the quantiza-
tion axis. There is no change in the saturation of the system and as a
consequence no change in the absorption.
On the other hand, $\Delta M=\pm 1$ transitions are induced by a laser beam when
the external static field is orthogonal to the EM field or is oriented
along the beam.
In this case the $\Delta M=-1$ and $\Delta M=+1$ transitions saturate independently
when the degeneracy is removed and a change in absorption is expected.
It is worth noting that in the case of an inhomogeneous line whose M
sublevels are resolved with respect to the homogeneous width the $\Delta M=+1$
transitions starting from a given M sublevel involve molecules with dif
ferent velocity components along the beam direction, i.e. different mo-
lecules. Again the saturated absorption coefficient can be computed as
a function of J and ΔJ selection rule by using the Clebsh-Gordan coef-
ficients

$$k^F(S) = \sum_{-J}^{+J} \left(k_M^+(S) + k_M^-(S) \right) = \sum_{-J}^{+J} \left(\frac{\alpha |\mu_M^+|^2}{\sqrt{1+\sigma|\mu_M^+|^2}} + \frac{\alpha |\mu_M^-|^2}{\sqrt{1+\sigma|\mu_M^-|^2}} \right) \quad 5)$$

Analytical expressions for $R(S)=k^F(S)/k(S)$ can than be given for any
J and ΔJ. In the case of a plane wave such equations were reported in
/6/ and /8/. In the case of a Gaussian TEM_{00} beam an analytical expres
sion for $R(S)$ can again be given for an optically thin sample as a
function of S_0, the saturation corresponding to the maximum intensity
of the light beam $(I(r)=I_0 \exp - (r/w)^2)$

$$R_Q(J,S_0) = \frac{2 \sum_{-J}^{+J} \left(\sqrt{1 + \frac{3}{4} S_0 \frac{J(J+1)-M(M+1)}{J(J+1)}} - 1 \right)}{\sum_{-J}^{+J} \left(\sqrt{1 + 3 S_0 \frac{M^2}{J(J+1)}} - 1 \right)} \quad 6)$$

$$R_R(J,S_0) = R_P(J+1,S_0) = \frac{2 \sum_{-J}^{+J} \left(\sqrt{1 + \frac{3}{2} S_0 \frac{(J+M)^2+3(J+M)+2}{(2J+1)(2J+3)}} - 1 \right)}{\sum_{-J}^{+J} \left(\sqrt{1 + 6 S_0 \frac{(J+1)^2 - M^2}{(2J+1)(2J+3)}} - 1 \right)} \quad 7)$$

where Q,R and P means a $J \leftrightarrow J$, $J \leftrightarrow J+1$, and $J \leftrightarrow J-1$ transition re-
spectively. In fig. 2 the computed results are shown both for integer
and half integer values of J. We found the same maximum absorption in-
crease $(R = \sqrt{2})$ for Q_1, R_0, $R_{\frac{1}{2}}$, and P_1 transitions. By increasing the J

values, the Q_J and R_J lines show a rapid convergence to two limiting
curves: $R_Q(J,S)$ increase with J and S up to a maximum value of 1.57,
while $R_R(J,S) = R_P(J+1,S)$ decreases to an asymptotic value of 1.27.
In both cases the effect is large and easily detectable. Only for the
$J=1/2 \leftrightarrow 1/2$ transition is $R_Q(1/2,S)\equiv 1$ as expected.

As for the shape of the NLHE as a function of the applied static field
in the general case of a $J \leftrightarrow J'$ transition it is worth to note that
the degeneracy may be removed in the upper and lower state at a diffe-
rent rate depending from their indetermination and the size of the
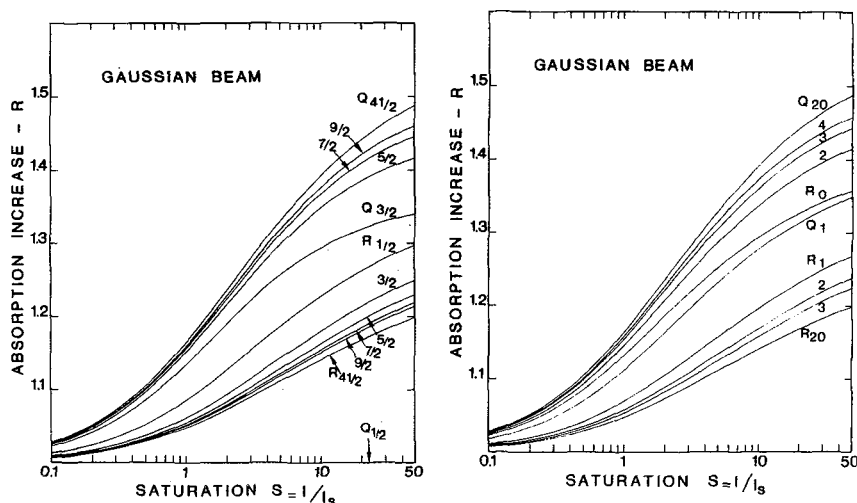Zeeman or Stark effect. As a consequence if the Zeeman or Stark effects

Fig. 2

are linear in the field and no HFS splitting exists within the Doppler
width the NLHE is expected to consist, at the first order, of two Lo-
rentzians with different width and amplitude. In many cases the two
Lorentzians will have nearly the same width. In this case the NLHE has
a Lorentzian shape.
The limiting case of high J transitions was investigated by using CO_2
laser excitation of rotovibrational transitions of polar molecules
exibiting a linear Stark effect /5,7/. In fig. 3 is shown the NLHE for
two transitions in CH_3OH detected by means of the optoacoustic effect.
As expected the NLHE is significantly smaller for the R_{26} line than
for the Q_{16} for about the same CO_2 laser power and gas pressure.
A general expression for the saturation intensity I_S cannot be given
since it depends on the relaxation processes of any specific system
/3/. In the infrared, where the rotational collisional relaxation rate
is the dominant process, largely independent from J, I_S is proportional
to p^2, where p is the gas pressure. Also this behaviour was experimen-
tally verified /6/.
A quantitative analysis of the NLHE lineshape can be done for atomic
transitions in the visible, which offer low quantum numbers. Measure-
ments have been performed by using the optogalvanic technique in a hol-
low cathode discharge, which provides an easily access to the whole
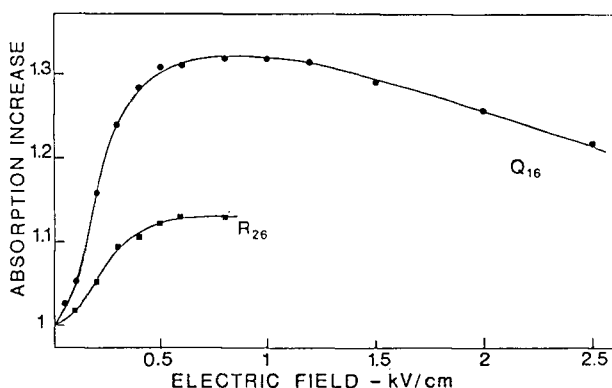spectra of both volatile and refractory elements. It was demonstrated

Fig. 3

that a magnetic field collinear with the laser beam and the discharge
does not perturb the signals or affect the S/N ratio.
Clearly resolved Zeeman pattern could be observed /9/.
An example of the NLHE as a function of the laser frequency and for a
fixed magnetic field is shown in fig. 4. The absorption increase all
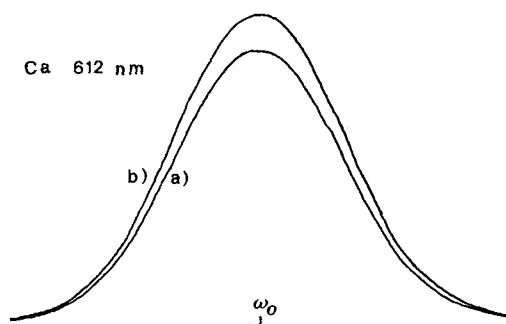over the Doppler profile is clearly demonstrated.

Fig. 4 - Doppler profile of the 612 nm Ca line.
Curve a: no field, curve b: 13 Gauss.

A quantitative analysis of the shape of the NLHE as a function of the
applied magnetic field will be published elsewere /10/. In fig. 5 is
shown the results for a $J=0\leftrightarrow1$ transition in Neon at 607.4 nm.
Since the g_S factor of the J=1 level is known a quantitative fit of the
experimental points to the theoretical curve composed of a Lorentzian
minus a Gaussian was possible /10/. The agreement was fairly good gi-
ving the following values for the independent parameters: $\Delta\nu_D=2160$MHz;
$\Delta\nu_H$ = 13.7 MHz; R=1.146. The Doppler width obtained directly by scan-
ning the line with a single mode Dye laser was found to be 2190(40)MHz.
The enhancement factor R, as obtained from the heigth of the Lorent-
zian, gives the saturation parameter which is used to evaluate the
contribution of the saturation to the homogeneous width. The indepen-
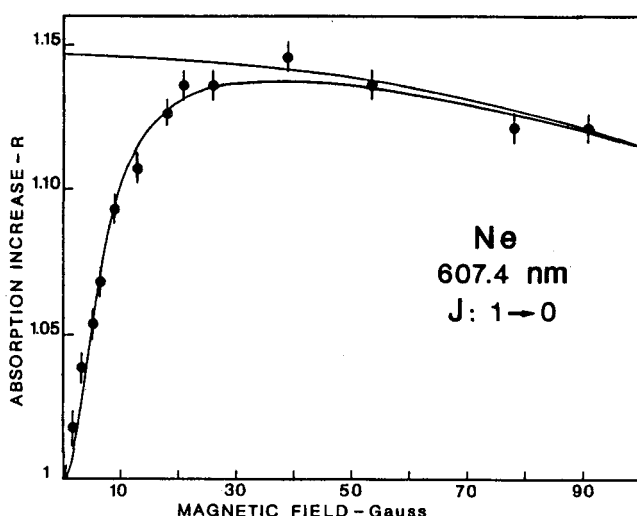dent determination of S_0 and $\Delta\nu_H$ is a unique advantage of the NLHE.

Fig. 5

The natural linewidth and the pressure broadening coefficient were then
found in agreement with existing data /10/.
For transitions with J=1↔1 or J=1↔2 a NLHE consisting of the sum of
two Lorentzians could also be observed leading to an independent de-
termination of the widths of the upper and lower levels /11/.

II - Application to the lasers output power

In the first part we have demonstrated that the absorption coefficient
of a transition is significantly increased as a consequence of the
NLHE. It is obvious that the same increase is expected in the satura-
ted gain of the laser medium when the population between the upper and
laser level is inverted.
As a consequence of the NLHE the output power of the gas laser is ex-
pected to increase when an external field is applied to the active me-
dium. In fact such an effect was early observed in He-Ne laser on the
0.63,1.15,and 3.39μm lines /12/. However all this laser lines are J =
= 1↔2 transitions and the relative intensity of the effect as a func-
tion of J as predicted by eqs. 6 and 7 cannot be tested. Moreover the
output power of the laser in this case is perturbed by conpetition
among the different transitions, which is also depending on the pre-
sence of an external field/12/.
Two different cases will be considered here that would permit a better
test of the predictions of the NLHE.

  - Optically pumped far-infrared lasers

The far-infrared (FIR) laser emission occurs between rotational tran-
sitions of polar molecules optically pumped in an excited roto-vibra-
tional state by means of resonant absorption of the powerful CO_2 laser
radiation. The pumping scheme is shown in fig.6. The pumping transi-
tion is Doppler broadened and strongly saturated while the FIR lasing

transition is homogeneous (pressure broadening). The absence of a discharge allows an easy application of an Electric field orthogonal to the laser axis and the linear Stark effect removes the degeneracy between the M sublevels. Hybrid waveguides must be used that force the FIR field to be orthogonal to the static Stark field. Hence only $\Delta M = \pm 1$ selection rules are allowed for the FIR transition, while either $\Delta M = \overline{0}$ or $\Delta M = \pm 1$ selection rules allowed for the pumping transition depending from the relative polarization of the CO_2 laser beam /13/.
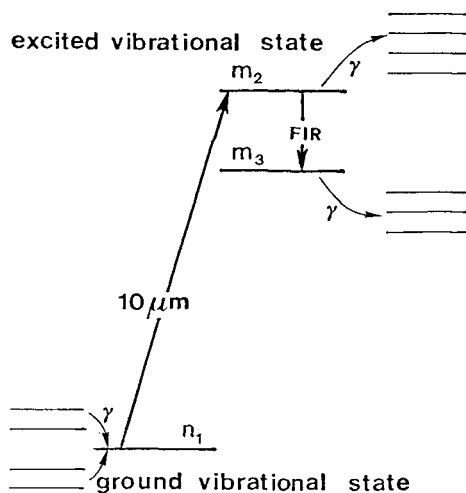

Fig. 6

For the $\Delta M = \pm 1$ pump selection rules an increase in the pumping rate is expected as a consequence of the NLHE. On the contrary the Stark effect on the FIR transition splits the line in many Stark components of which only one can be put in resonance with a FIR cavity mode. As a consequence only a gain decrease is expected on the FIR transition. In fig. 7 are reported the experimental results /7/ obtained for FIR laser lines of about the same efficiency and wavelength.
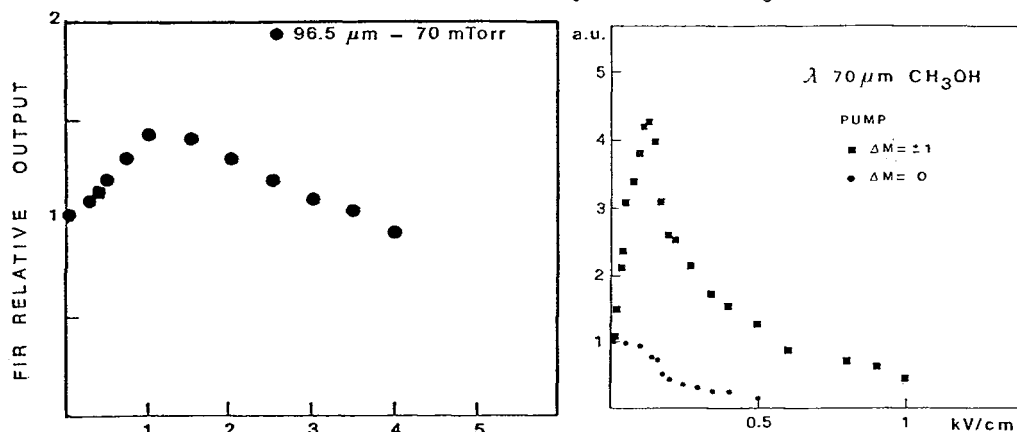

Fig. 7

The pump transition is respectively a Q_9 line for the 70 μm laser and a R_{26} for the 96 μm laser. As a consequence the NLHE and the output power increase was found to be larger for the 70 μm line as predicted. It is also shown that in the case of $\Delta M=0$ pump selection rule the output power showed only a power decrease, again as predicted. This result proves that the NLHE enhancement is in the pump transition since the polarization of the 70 μm line is always orthogonal to the Stark field.

Argon and Kripton ions laser

This lasers are the most powerful sources of CW radiation in the visible and are produced in large number. All the commercial models are supplied with a solenoid that produces an axial magnetic field of about 0.1 T on the discharge tube. The output power is as a consequence increased by a factor up to four.
The reason of this behaviour is not properly understood up to now.
In standard textbooks /13/ and in extensive reviews of the subject /14--16/ it is tentatively assumed that the output power enhancement may be due to an increase in the electron density in the plasma at given value of the current. However experimental investigations gave contradictory results on this points /14/. Moreover an electron density increase would produce about the same enhancement effets on all the visible laser lines. On the contrary the NLHE predics a different enhancement as a function of J and ΔJ independent of the current saturation effect. In particular no effect is predicted for the $J=1/2\leftrightarrow1/2$ laser lines. Unfortunately the available experimental data were obtained in multimode operation and only a qualitative comparison with the theory is possible. Moreover the homogeneous linewidth of the ion lasers is quite large (500∿800 MHz) as a consequence of the short lifetime of the lower levels and the ratio $\Delta\nu_D/\Delta\nu_H$ is about 6∿7.

In fig. 8 is shown the predicted relative gain increase R(J,S) for all the CW laser lines of Ar and Kr ion lasers as a function of the saturation parameter S_0. The validity of eq. 1 has been assumed. In fact an exponent larger than 1/2 may be possible as a consequence of the low $\Delta\nu_D/\Delta\nu_H$ ratio that would lead to larger R(S) values.
In fig. 9 are shown experimental result obtained from the Hughes report /17/. The NLHE predicts no enhancement for the 458 nm laser lines and the small experimental effect may be due to the expected electron density increase. A similar effect has been observed in Kr^+ red lines. The 647 nm line is a $J=3/2\leftrightarrow5/2$ transition, while the 676 nm is $J=1/2\leftrightarrow1/2$. The first shows a large power increase as a function of the field, while the second one is practically insensible to the field. The relative enhancements of the 476 nm and 488 nm laser lines is also in agreement with the prediction. The theory predicts also an equal enhancement for both the 488 nm and the 514 nm laser lines. The experimental results apparently are in disagreement, the enhancement of the 514 nm line being larger. However the saturation intensity I_s of this line is of about 50 W/cm^2 ten time larger than that of the 488 and 476 nm lines. The output power is nearly the same for the 488 and 514 lines and as a consequence the actual value of S_0 is about one order of magnitude smaller and of the order of unity for the second one. The NLHE cause a positive feedback loops: the field increases the saturated gain which increase the laser beam intensity which increases the gain and so on. As a con-

sequence the relative power increase is expected to be larger for low S_0 value where the slope of R(S) is larger.
As for the optimum value of the magnetic field (800∿1000 Gauss), it is also in agreement with the prediction of eq.3 if we consider the large value of the homogeneous line.
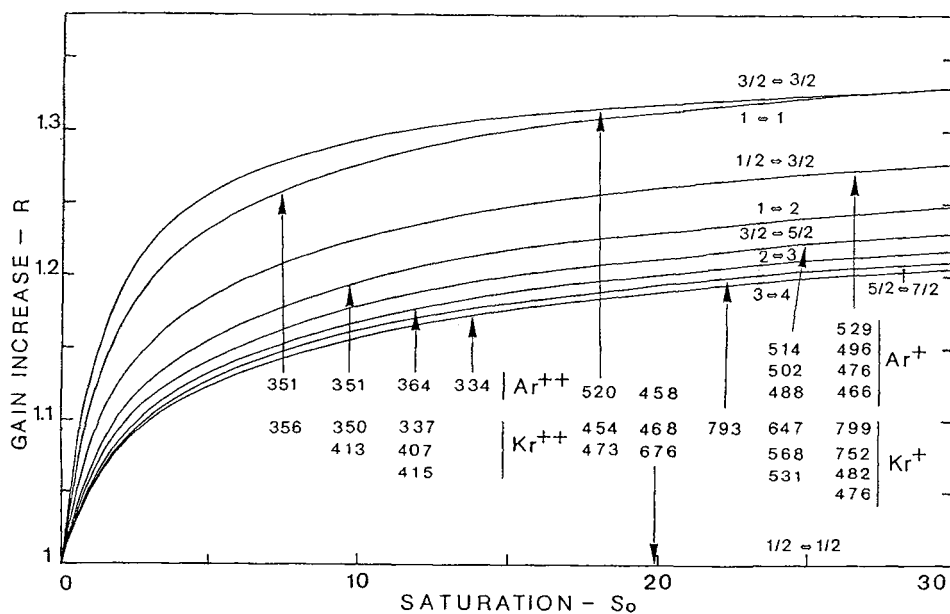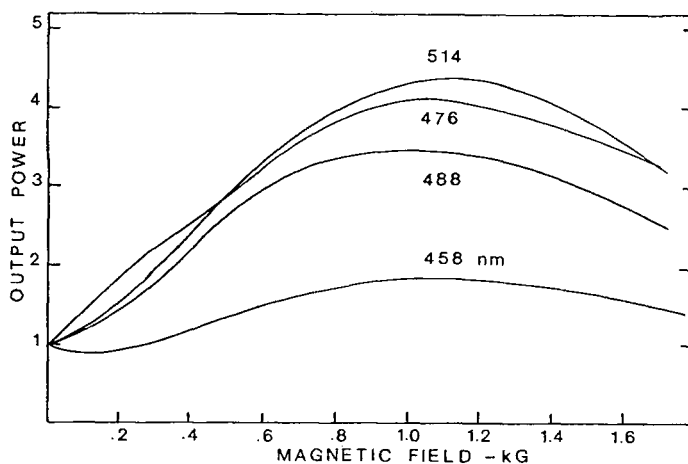

Fig. 8

Fig. 9

In conclusion the NLHE predictions are in qualitative agreement with the experimental results. Quantitative comparisons can be performed only with the experimental determinations of the saturated gain increase in single mode operation. At present the NLHE is however the most straight forward interpretation of the power increase in noble gases ion laser as a function of the axial magnetic field.

References

1 - W. Hanle: Naturviss. $\underline{11}$, 691 (1923)

2 - A.C. Luntz, R.G. Brewer, K.L. Foster, J.D. Swalen: Phys .Rev.Lett. $\underline{23}$, 951 (1969)

3 - A. Corney: "Atomic and Laser Spectroscopy" Clarendon Press, Oxford 1977.

4 - M.S. Feld, A. Sanchez, A. Javan, B.J. Feldman: Publ. .n.217 du CNRS - Paris 1974 pp. 87-104

5 - F. Strumia, M. Inguscio, A. Moretti: in "Laser Spectroscopy V" A.R.W. McKellar, T.Oka, B.P. Stoicheff (Eds.) Springer, Berlin Heidelberg, New York 1981, pp.255

6 - M. Inguscio, A.Moretti, F.Strumia: Appl. Phys. $\underline{B28}$, 88 (1982)

7 - M. Inguscio, A.Moretti, F.Strumia: IEEE J.Quant.Electron. QE-16, 955 (1980)

8 - F. Strumia: in "Advances in Laser Spectroscopy" F.T.Arecchi, F.Strumia, H.Walther (eds.) - Plenum, New York 1983

9 - N.Beverini, M.Galli, M.Inguscio, F.Strumia, G.Bionducci: Opt. Commun. $\underline{43}$, 261 (1982)

10 - N. Beverini, M. Inguscio, F.Strumia: Appl. Phys., in press.

11 - B. Barbieri, N.Beverini, G.Bionducci, M.Galli, M.Inguscio, F.Strumia: in "Laser Spectroscopy VI", H.P.Weber, M.Luthy(eds.) Springer, Berlin, New York, 1983

12 - M.I. Dyakonov, S.A. Fridrikhov: Soviet Phys. Uspekhi, $\underline{9}$, 837 (1967) and references therein.

13 - F. Strumia, M.Inguscio: in "Infrared and millimeter waves" vol. 5 K.J. Button (ed.) Academic Press, New York 1982, pp.129-213

14 - C.C. Davis, T.A. King: in "Advances in Quantum Electronics" vol.3 D.W. Goodwin (ed.) Academic Press, New York 1975, pp.169-454

15 - W.B. Bridges: in "Methods of experimental physics" vol. 15A C.L. Tang (ed.) Academic Press, New York 1979 pp.33-166

16 - M.H. Dunn, J.N. Ross: in "Progress in quantum electronics" vol.4 J.H. Sandars, S. Stenholm (eds)Pergamon Press, Oxford, New York 1977, pp.233-264

17 - W.Bridges, A.S.Halsted: "Gaseous Ion Laser Research"Fin.Tech.Rep. Hughes Res.Lab.(1967)(unpublished)NTIS access numb.AD-814897.