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Optogalvanic sub-Doppler spectroscopy in titanium

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Abstract. The a $^3P_0$–y $^3D_1^o$ Ti I transition at 592.2 nm was investigated by using a hollow-cathode discharge. Sub-Doppler resolution was obtained by combining optogalvanic detection with saturation spectroscopy and the non-linear Hanle effect. The upper state Landé factor was also determined by Zeeman intermodulated spectroscopy.

1. Introduction

The combination of optogalvanic detection with Doppler-free techniques has proved in many cases to be well suited to achieve high resolution (Barbieri *et al* 1990). Since the advent of tunable dye lasers optogalvanic detection has been widely used as an alternative spectroscopic technique in spite of the complexibility of a theoretical description of the optogalvanic effect. Optogalvanic spectroscopy (as well as optoacoustic) is sometimes referred to as an unconventional technique in contrast to most of other spectroscopy techniques where scattered light is detected (Ernst and Inguscio 1988). Optogalvanic detection is in particular very convenient for the study of non-volatile elements which can be easily detected and studied when sputtered in discharges. In addition the simplicity of its experimental set-up and its great sensibility makes this technique attractive for several applications.

Recently we reported Doppler-free spectra of the a $^3P_0$–y $^3D_1^o$ Ti I transition, at 592.2 nm, obtained by intermodulated optogalvanic spectroscopy (IMOGS) in a hollow-cathode lamp (Cruz *et al* 1994). The wavenumber of this transition was determined relative to iodine lines and the isotope shifts were also obtained using a natural abundance sample.

In the present work we extend our sub-Doppler spectroscopic investigation of titanium with a focus on the reliability of optogalvanic detection to perform high resolution spectroscopy. For this we used different Doppler-free techniques combined with optogalvanic detection. Linewidth measurements of the a $^3P_0$–y $^3D_1^o$ Ti I transition obtained from these techniques are compared. The homogeneous linewidth was obtained from the intermodulated spectra (Cruz *et al* 1994) by fitting them to theoretical expressions which describe the saturation spectroscopy lineshapes under the influence of velocity-changing collisions (Smith and Hansch 1971, Tenenbaum *et al* 1983). Doppler-free optogalvanic spectra were also obtained by using a conventional saturated

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spectroscopy set-up where a dip is created in the centre of the Doppler profile (Lamb dip) (Demtröder 1988). Dips associated with the less abundant titanium isotopes are also observed with this set-up although with a lower signal-to-noise ratio than in the intermodulated spectra (Cruz *et al* 1994).

Zeeman IMOGS (Beverini *et al* 1982) was performed by applying a longitudinal magnetic field. The upper state Landé factor was then determined by measuring the splittings of the Zeeman sublevels as a function of the magnetic field.

The a $^3P_0$–y $^3D_1^o$ transition of titanium is also convenient for a study of the non-linear Hanle effect (NLHE) since its theoretical analysis is simpler for a $J''=0$–$J'=1$ transition (Moruzzi *et al* 1991). This transition has also good intensity and falls within the tuning range of a dye laser operating with Rhodamine 6G. By considering a longitudinal magnetic field and linear polarization of the laser beam a $J''=0-J'=1$ transition actually behaves as a V-type level system. The linear polarization of the laser may be considered as a superposition of a $\sigma-$ and $\sigma+$ polarizations which then induce transitions from the lower level to the upper $m=-1$ and $m=+1$ sublevels respectively. With this configuration the upper $m=0$ sublevel may be neglected. The non-linear absorption coefficient changes in the transition from a two-level system ($B=0$) to a V-type level system ($B>0$) and the optogalvanic signal can be used to monitor this change.

Non-linear Hanle effect curves (optogalvanic signal plotted against magnetic field) were obtained for several values of laser intensities and current in the discharge and the results are compared to those obtained from the saturated spectra.

2. Experimental details

The construction details of a homemade hollow-cathode lamp have been previously described (Mirage *et al* 1992). We used a 20 cm long glass tube with diameter of 4 cm. The lamp was sealed with argon (buffer gas) at a pressure of 1 Torr and could be operated at current values up to 150 mA. It has double anodes and a titanium cathode, this being a 10 mm long cylindric tube with internal diameter of 3 mm.

As a source of radiation we used a commercial single-frequency ring dye laser with a linewidth of about 1 MHz. The laser polarization was linear and the beam was Gaussian with a diameter which nearly matched the cathode one. The laser positioning and tuning procedure as well as the set-up for IMOGS were described previously (Cruz *et al* 1994). Laser power values of typically a few tens of mW in each counterpropagating beam were used in obtaining the intermodulated spectra. They are affected by some amount of power broadening which could be taken into account by extrapolating several measurements to null laser power. Lower laser power levels resulted in worse signal-to-noise ratios specially at low current values in the discharge (less than 40 mA). Doppler-free optogalvanic spectra were also obtained by using a saturated absorption set-up where a weak beam was modulated by a mechanical chopper and a strong one was sent counterpropagating. Using a lock-in the optogalvanic signal was detected at the modulation frequency of the weak beam. In this way we obtain the Doppler profile of the transition with a dip at the central frequency (Lamb dip) (Demtröder 1988). As is well known the width of this dip is the homogeneous width of the transition and its height depends on the saturation parameter. These spectra were also recorded at several values of laser power and current in the discharge. The linearity of the laser scanning was monitored by recording all sub-Doppler spectra simultaneously with the transmission peaks of a Fabry–Perot interferometer with a free spectral range of 75 MHz.

The application of a longitudinal magnetic field allowed us to perform Zeeman IMOGS (Beverini *et al* 1982). For this we constructed a solenoid in which the lamp was inserted. A magnetic field up to 350 G could be produced with good homogeneity in the region where the lamp was positioned. The same solenoid was used in the NLHE measurements. In these measurements we positioned the laser frequency in the maximum of the Doppler profile and the optogalvanic signal was recorded as a function of the magnetic field. This was also done for several values of laser intensity and current in the lamp. We did not use lenses to focus the beam into the lamp since it caused a decrease in the magnitude of the optogalvanic signal (OGS). However we observed that the NLHE curve shapes were independent of focusing of the laser beam. They are also independent on the laser frequency position inside the Doppler profile (Moruzzi *et al* 1991). By using lamps sealed at different pressures we observed that the magnetic field could be applied only within a certain pressure range without causing instabilities in the OGS. A pressure of about 1 Torr was experimentally determined by us as convenient for the measurements when a magnetic field was used. These instabilities were observed for pressures higher than 1.5 Torr. We observed that the configuration of double anodes also played an important role in avoiding such instabilities. We used homemade lamps with only one anode which were less stable when a magnetic field was applied. Measurements with different techniques were performed with the same lamps.

3. Results and discussion

Figure 1 shows a typical intermodulated spectrum of the a $^3P_0$–y $^3D_1^o$ Ti I transition obtained at a current of 100 mA. A large Gaussian pedestal due to velocity-changing

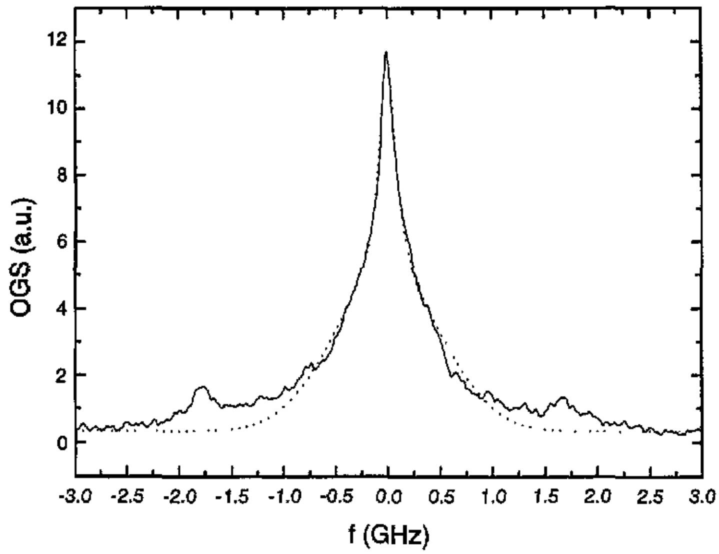

Figure 1. Sub-Doppler spectrum of the $^3P_0$–$^3D_1^o$ transition of Ti I obtained by intermodulated optogalvanic spectroscopy (the current in the hollow-cathode lamp was 100 mA). The three peaks correspond to isotopes 46, 48 and 50 (Cruz *et al* 1994). The broken curve is the fit of equation (1) to the spectrum. The 'zero' position in the horizontal axis is arbitrary.

collisions is observed. The strong peak corresponds to the most abundant isotope (^{48}Ti, 73.8% natural abundance) and the two lateral peaks correspond to the isotopes 46 (8%) and 50 (5.4%) (Cruz *et al* 1994). The homogeneous linewidth was obtained from the intermodulated spectra recorded at several values of laser power and current in the lamp by considering theoretical expressions which describe the lineshapes in saturated spectroscopy by taking into account the influence of velocity-changing collisions (Tenenbaum *et al* 1983). These expressions were obtained for the limiting cases of 'strong' and 'weak' collisions and our intermodulated spectra were better fitted to the case of strong collisions. In this case the lineshapes consist of a Lorentzian profile superimposed onto a Gaussian pedestal. The adjusted expression was (Tenenbaum *et al* 1983):

$$S(\Delta\nu)=\frac{\sigma_0\gamma_0 d}{2\sqrt{\pi}\Delta\nu_D}\left(\frac{(\frac{1}{2}\gamma_0)^2}{(\frac{1}{2}\gamma_0)^2+\Delta\nu^2}+C\exp[-(\Delta\nu/\Delta\nu_D)^2]\right)\times\frac{N\exp[-(\Delta\nu/\Delta\nu_D)^2]}{1+D+C\exp[-(\Delta\nu/\Delta\nu_D)^2]} \tag{1}$$

where σ_0 is the resonant cross section, γ_0 is the transition homogeneous linewidth, d is the absorption length, $\Delta\nu_D$ is the Doppler width, C is a parameter which specifies the relative contribution of the Gaussian and Lorentzian terms and D gives the non-linear dependence of the lineshape on the pump intensity and collisions; N is the total atomic density. The broken curve in figure 1 is the fit of equation (1) to the experimental spectrum.

In the fits we fixed $\Delta\nu_D=960$ MHz (FWHM = 1600 MHz), obtained by fitting three Gaussians to the peaks of the isotopes 46, 48 and 50 in the Doppler broadened spectra (Cruz *et al* 1994). By considering the laser power values used to obtain the intermodulated spectra we consider the case of strong pumping and fixed $D=0$ (Tenenbaum *et al* 1983). The adjusted parameters were γ_0 and C. The other parameters may be considered as only one amplitude factor. The values obtained for γ_0 at different laser power were extrapolated to null power and the obtained value for γ_0 was 140 ± 20 MHz (FWHM) where the quoted error is one standard deviation.

Figure 2 shows intermodulated spectra obtained at different values of current in the lamp. It is observed a decrease (with increasing current) in the Gaussian pedestal relative to the homogeneous Lorentzian signal. The change in the magnitude of the strong peak is characteristic of the dependence of the OGS on current for this transition. For fixed laser power values the OGS showed an increase up to 80 mA and then a slow decrease up to 150 mA. By fitting equation (1) to all the intermodulated spectra obtained at different current values it was observed a decrease of the parameter C with current increase. By plotting the inverse of these fitted parameters against the current in the lamp we obtain a straight line with a slope equal to 1.1×10^{-2} mA^{-1}. This behaviour is similar to that observed by Sasso *et al* (1988) for neon and is attributed to a decrease, with current increase, of the lower level effective lifetime due to collisions with electrons which in turn decreases the time available for the atoms undergo velocity-changing collisions (Sasso *et al* 1988).

Figure 3 shows saturated absorption spectra obtained by detection of the OGS at the modulation frequency of a weak beam in the presence of a strong counterpropagating one (Demtröder 1988). With this set-up we observe the Doppler profile of the transition with a dip at the zero velocity group (Lamb dip) when the atoms interact simultaneously with both the weak and strong beams. In figure 3 the upper curve was

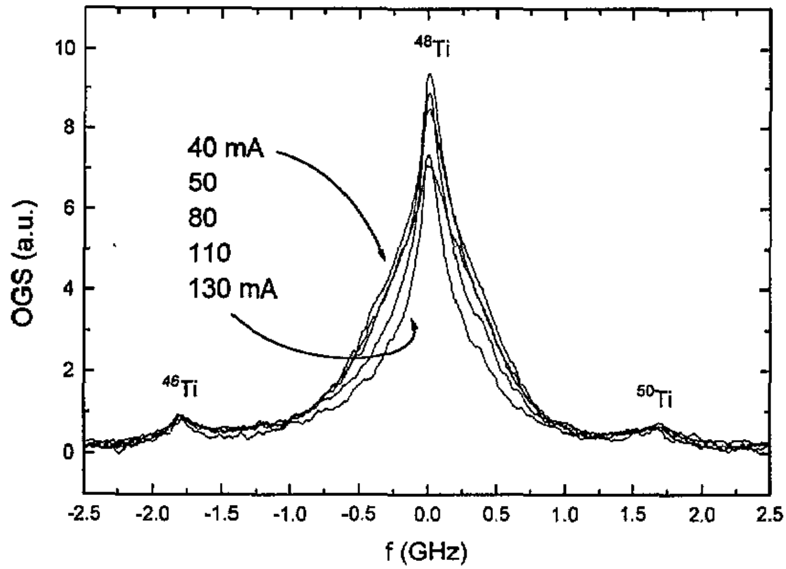

Figure 2. Variation with lamp current of the Doppler pedestal in intermodulated optogalvanic spectra. A plot of $1/C$ (C is the fitted parameter in equation (1)) against current *gives a straight line with a slope equal to* 1.1×10^{-2}*/mA.*

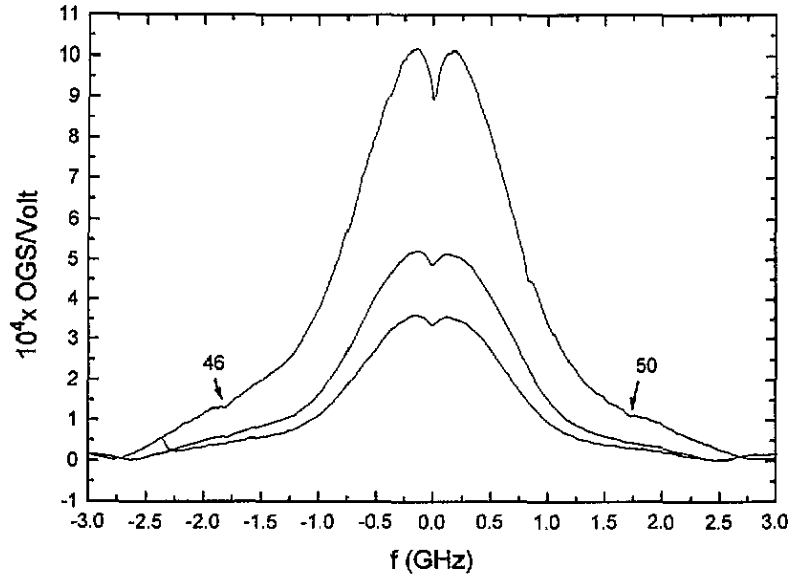

Figure 3. Optogalvanic detection of the Lamb dip in the $^3P_0$–$^3D_1^o$ Ti I transition. The upper curve was obtained at $i = 80$ mA. The intermediate one was obtained at the same laser power and at $i = 35$ mA. The lower curve was obtained at $i = 35$ mA and at a lower laser power. The arrows indicate the dips associated with the less abundant even isotopes.

obtained at a current of 80 mA. The intermediate spectrum was obtained with the same laser power but with a current of 35 mA. The lower spectrum was also obtained at 35 mA but with a smaller value of laser power. The dips corresponding to the isotopes 46 and 50 are observable in the spectrum obtained at 80 mA but with a smaller signal-

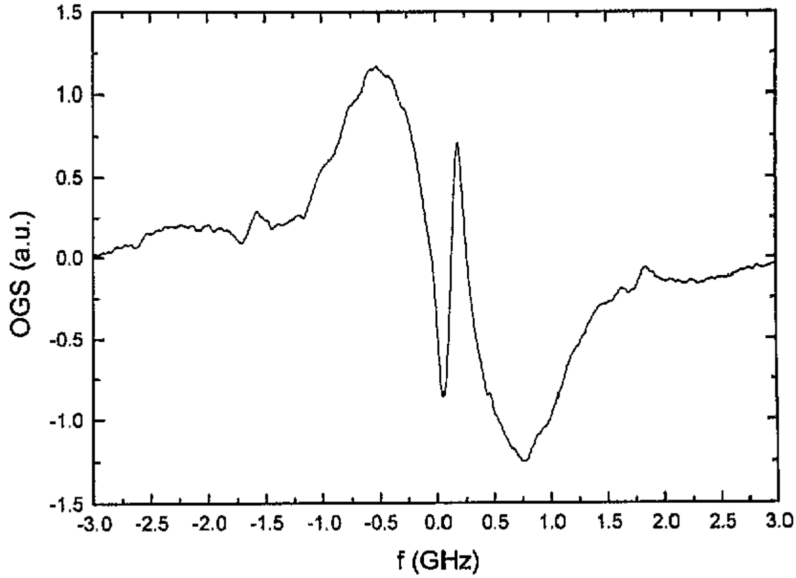

Figure 4. First derivative optogalvanic spectrum of the $^3P_0$–$^3D_1^o$ Ti I transition obtained with laser frequency modulation and the same set-up as figure 3. The peaks corresponding to isotopes 46 and 50 are also observable.

to-noise ratio than in the spectra obtained by IMOGS (figure 1). The width of the saturation dips is the homogeneous width of the transition (including power broadening) and its height depends on the saturation parameter. Figure 4 shows a spectrum obtained with the same experimental configuration but with frequency modulation of the laser and the lock-in referenced to this modulation frequency. This curve may be useful for laser frequency stabilization to the centre of the transition using the OGS. The spectra obtained in this way are the first derivative of those of figure 2. The peaks corresponding to the isotopes 46 and 50 are also observed in figure 4. By measuring the width of the dip observed in these spectra (figures 3 and 4) and extrapolating the values to null laser power we obtained $\gamma_0 = 120 \pm 10$ MHz (FWHM) (the quoted error is one standard deviation) which agrees (as expected) with the value obtained from the intermodulated spectra. The value of 130 ± 20 MHz for the homogeneous linewidth (FWHM) obtained from the saturated absorption spectra (intermodulated and Lamb dip) is expected to be dominated by collisional broadening.

Figure 5 shows spectra obtained by Zeeman IMOGS (Beverini *et al* 1982) at different values of the external magnetic field and current in the lamp. The laser polarization is linear and the magnetic field is applied along the lamp axis. With this configuration we observe for the isotope 48 two lateral peaks (in the centre of figure 5) which correspond to transitions between the lower level and the $m = -1$ and $m = +1$ upper Zeeman sublevels. The central peak is a crossover resonance which appears because both transitions share the lower level. In this way Zeeman spectroscopy was carried out combined with a Doppler-free technique. The application of a magnetic field did not prevent performing high resolution spectroscopy in a hollow-cathode discharge. Figure 6 shows the splitting of the upper Zeeman sublevels for the isotope 48 at several values of the magnetic field. All curves were recorded at 110 mA and a fixed value of the laser power. From these measurements we obtained the following value for the upper state Landé factor: $g_{J'} =$

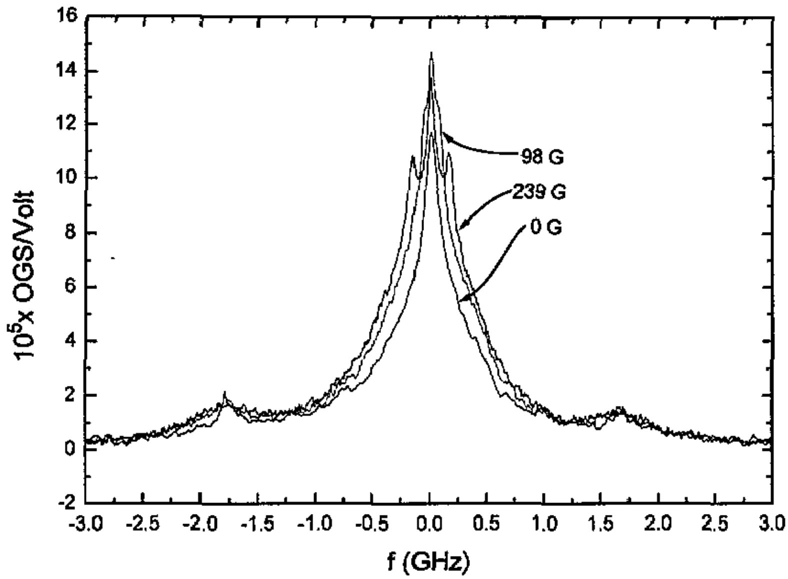

Figure 5. Spectra of the $^3P_0$–$^3D_1^o$ Ti I transition obtained by Zeeman IMOGS at $i = 100$ mA and $B = 0$, 98 and 239 G. The lateral peaks near $f = 0$ correspond to transitions between the lower level and the $m = -1$ and $m = +1$ upper Zeeman sublevels. The central one is a crossover resonance.

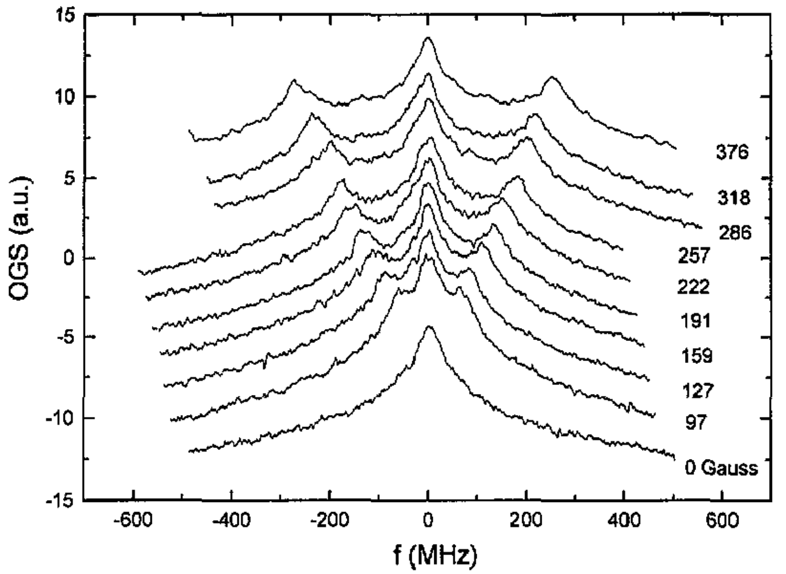

Figure 6. Zeeman intermodulated optogalvanic spectra showing the splitting in the upper state of the $^3P_0$–$^3D_1^o$ transition of Ti I ($i = 110$ mA). The central peak is a crossover resonance. A plot of the Zeeman splittings against magnetic field allowed the determination of the Landé factor of the level $^3D_1^o$.

0.49 ± 0.02 where the quoted error is one standard deviation. This value agrees with the tabulated one (Moore 1971). We point out that an independent determination of Landé factors, when they are not known, is necessary for the analysis of the NLHE results.

In the measurements of the NLHE we monitor the saturated absorption coefficient of a sample as a function of an external field. Let us consider a $J''=0$–$J'=1$ transition with a Doppler width ($\Delta\nu_D$) much larger than the homogeneous width (γ_0) (Doppler limit), an external longitudinal magnetic field and linear polarization of a single-frequency laser beam (with a linewidth smaller than the transition homogeneous linewidth). By tuning the laser frequency to the maximum of the Doppler profile we can think that initially (when $B=0$) the laser interacts with the zero velocity group of atoms. As the magnetic field is applied an absorption increase is observed (which may be considered nearly Lorentzian (Moruzzi *et al* 1991)) which occurs in a field region where the upper state Zeeman sublevels are still superimposed within their homogeneous linewidths (collisional and power broadened). The magnitude of this increase depends on the intensity of the laser. For this magnetic field interval the atoms are then in a coherent superposition of the $m=-1$ and $m=+1$ upper Zeeman sublevels (Moruzzi *et al* 1991). The transition from a two-level system (when $B=0$) to a three-level system (when $B>\gamma_u h/g_j\mu$ where γ_u and g_j are the upper level width and Landé factor, respectively, h is the Planck constant and μ is the Bohr magneton) leads to a change in the saturation parameter and then in the non-linear absorption coefficient. As the magnetic field increases the $m=-1$ and $m=+1$ sublevels will be completely split and the laser (kept fixed in frequency) will no longer be in resonance with the atoms. However for an inhomogeneously (Doppler) broadened transition the laser will then start to interact with atoms at other velocity groups, symmetrically displaced in the Doppler profile, and which were not in resonance when $B=0$. As the magnetic field is increased further the absorption coefficient will then decrease following the Gaussian velocity distribution of the atoms (Moruzzi *et al* 1991). Therefore from NLHE curves for transitions where $\Delta\nu_D\gg\gamma_0$ one can obtain the homogeneous linewidth from the absorption increase at small B values (small enough to keep the Zeeman sublevels overlapped) and the Doppler width from the absorption decrease which is observed at larger B values. One feature of this technique is that it allows the determination of the individual width of the levels involved in a transition instead of the transition homogeneous linewidth as in most of the Doppler-free techniques. A $J''=0$–$J'=1$ transition is particularly suitable for this purpose since only the upper level is split by the magnetic field. By subtracting the individual level width obtained by NLHE from the homogeneous linewidth obtained with some sub-Doppler technique we can determine the width of the other level.

We intended to test the reliability of the optogalvanic detection as a way of monitoring the saturated or non-linear absorption coefficient in a NLHE experiment on the a $^3P_0$–y $^3D_1^o$ Ti I transition.

Figure 7 shows NLHE curves (optogalvanic signal plotted against longitudinal magnetic field) for the $^3P_0$–$^3D_1^o$ Ti I transition obtained at a current of 40 mA and different values of laser power. All curves obtained at different values of current and laser power were normalized to the value of the optogalvanic signal at $B=0$. In figure 7 is observed an increase in the optogalvanic signal with magnetic field increase which corresponds to the removal of degeneracy in the upper $J=1$ level (Moruzzi *et al* 1991, Beverini *et al* 1985). As expected this increase is followed by a decrease for higher magnetic fields corresponding to the scanning of the Doppler profile. A function which is the difference between a Gaussian and a Lorentzian centred at $B=0$ was adjusted to the experimental normalized NLHE curves (Moruzzi *et al* 1991). The fitted widths of the Lorentzian and Gaussian functions, in magnetic field units, were then transformed to frequency units by the relation $h\Delta\nu$ (FWHM) $=2g_J\mu B_{HM}$, where B_{HM} is the fitted half width at half maximum of both functions (Beverini *et al* 1985). The Lorentzian gives the width of

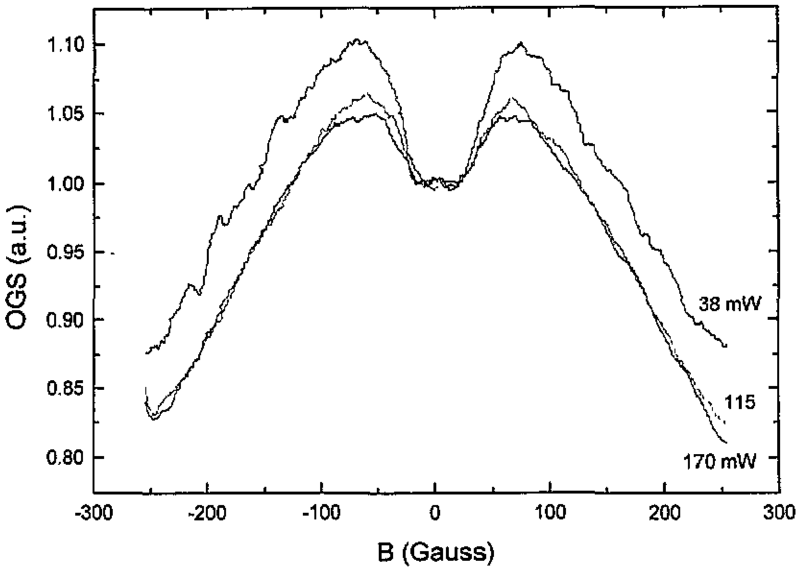

Figure 7. Optogalvanic signal as a function of a longitudinal magnetic field at laser power equal to 38, 115 and 170 mW showing the NLHE on the upper level of the $^3P_0$–$^3D_1^o$ Ti I transition. The laser polarization was linear and the current in the lamp was 40 mA.

the upper level and the Gaussian gives the Doppler width. The flat feature around $B = 0$ in figure 7 was due to the current source for the magnetic field and was properly taken into account in the fits.

In our measurements the signal increase has a strong dependence on the laser intensity as expected from a saturation effect. However this dependence is not as expected (Moruzzi *et al* 1991, Beverini *et al* 1985). The normalized NLHE increase increases with laser power up to a certain value (≈ 40 mW) and then decreases for higher laser power values (figure 7).

We also observed a dependence of the magnitude of this increase on the current in the lamp. This result contrasts with those of Beverini *et al* (1985) obtained for calcium and neon with optogalvanic detection, where no current dependence was observed. In our measurements the width associated with the decrease at larger B values was also smaller than the Doppler width. The value obtained by fitting a Gaussian to this decrease varied with current in the lamp and was larger for larger current. However the largest value was about half of the Doppler width obtained by scanning the laser across the transition (Cruz *et al* 1994). In spite of this behaviour for the signal decrease the width associated with the increase due to the NLHE (figure 7) varied little with current, probably because this increase occurs for much lower magnetic fields. The same was also observed by us in NLHE measurements of Ar^+ transitions in hollow-cathode lamps (Cruz *et al* 1995). From the Lorentzian part of the fitted function we obtained $B_{HM} = 42 \pm 4$ G (the quoted error is one standard deviation of measurements obtained at different current and laser power values). This value corresponds to an upper state width (FWHM) of 58 ± 6 MHz including the contribution of power broadening. It showed a small variation with current in the lamp and would be reduced up to 45 ± 6 MHz if we take into account power broadening at the laser power levels used in our measurements. If we then consider it as a reliable estimation of the upper level width and note that $A_{ik}/2\pi = 0.54$ MHz (Wiese and Fuhr 1975), the obtained value essentially reflects the collisional broadening of the level at the fixed pressure of our measurements. Considering in addition the homogeneous linewidth obtained from the saturated spectra we would have a width of approximately 85 MHz for the lower level. Making the assumption that the collisional broadening is similar for both lower and upper levels, we then find that the upper level would have a larger radiative lifetime than the lower one. One possible explanation is that the upper level width measurement would be affected by radiation trapping but this hypothesis needs to be verified by measurements at different pressures.

We could think that the variation of the signal increase with current could be attributed to the change with current of the spatial distribution of the sputtered atoms. If the beam is not focused in the lamp and is supposed to be Gaussian then we could have different effective saturation parameters at different current values as atoms interacting with different parts of the beam would effectively contribute to the signal. However we did not observe a change in this behaviour when we focused the beam into the lamp, when we should not expect this effect.

We believe that the characteristics of our NLHE results are due to the behaviour of the hollow-cathode discharge (and not the optogalvanic signal) under the influence of a magnetic field. This hypothesis is supported by NLHE measurements on Ar^+ transitions in hollow-cathode lamps where we compare NLHE curve shapes obtained by optogalvanic and fluorescence detection (Cruz *et al* 1995). We should mention that for titanium we could not compare optogalvanic NLHE curves with those obtained by detecting the fluorescence or absorption due to the small signal-to-noise ratio for these quantities. This behaviour did not prevent us obtaining good results with optogalvanic detection when precise determination is required only for frequency intervals as in the case of the Landé factor measurement by Zeeman IMOGS.

We should point out that the comparison between widths obtained by saturation spectroscopy and NLHE is not dependent on the fitting procedures used to determine them.

4. Conclusion

We presented optogalvanic sub-Doppler spectroscopy results for the a $^3P_0$–y $^3D_1^o$ transition of titanium. This element was sputtered in a hollow-cathode discharge and studied with a single-frequency dye layer.

Sub-Doppler optogalvanic spectra were obtained by intermodulated optogalvanic spectroscopy (IMOGS) and a saturation spectroscopy set-up where the Lamb dip is observed. The intermodulated spectra were well fitted to theoretical expressions for the case of 'strong' velocity-changing collisions.

The application of a longitudinal magnetic field allowed us to perform Zeeman IMOGS. The upper state Landé factor was then determined for this Ti I transition and is in good agreement with the tabulated value.

The non-linear Hanle effect was also investigated for this $J''=0$–$J'=1$ titanium transition. The obtained curve shapes (optogalvanic signal plotted against magnetic field) are characterized by a signal increase for small magnetic fields and a decrease for larger B values but show an unexpected dependence on the current in the lamp in contrast to other results in the literature. The dependence of the magnitude of the signal increase on the saturation parameter was not in quantitative agreement with calculated results (Moruzzi *et al* 1991, Beverini *et al* 1985) and in addition the width associated with the signal decrease did not agree with the Doppler width obtained directly by scanning the laser across the transition. We believe that this quantitative disagreement is due to the behaviour of the hollow-cathode lamp when submitted to a magnetic field. In this case careful attention to this problem is needed in such experiments particularly because it may depend on the construction details of the hollow-cathode lamps.

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