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INTERMODULATED OPTOGALVANIC SPECTROSCOPY - A COMPARISON WITH OTHER HIGH RESOLUTION TECHNIQUES

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Résumé - Nous faisons une comparaison entre les spectroscopies de fluorescence intermodulée, optogalvanique intermodulée, POLINEX (excitation par polarisation intermodulée) et de polarisation comme méthodes de spectroscopie sans effet Doppler dans une décharge à cathode creuse. La discussion est principalement basée sur des expériences réalisées sur les transitions $2p^5 3s\ ^3P_2 - 2p^5 3p\ ^3P_1$ de Ne à λ=5882 Å et $4d^5 5s\ ^5S_2 -$ $4d^5 5p\ ^5P_3$ de Mo à λ=5506 Å.

Quelques aspects généraux concernant la cathode creuse comme instrument en spectroscopie laser à haute résolution sont aussi brièvement discutés.

Abstract - A comparison of intermodulated fluorescence spectroscopy, intermodulated optogalvanic spectroscopy, POLINEX (POLarization INtermodulated EXcitation) spectroscopy and polarization spectroscopy used in Doppler-free spectroscopy on hollow cathode discharges is made, mainly based on experiments performed on the $2p^5 3s\ ^3P_2 - 2p^5 3p\ ^3P_1$ transition in Ne at λ=5882 Å and the $4d^5 5s\ ^5S_2 - 4d^5 5p\ ^5P_3$ transition in Mo at λ=5506 Å. Some general aspects regarding the hollow cathode as a tool in high-resolution laser spectroscopy are also briefly discussed.

I - INTRODUCTION

The extension of the optogalvanic spectroscopy to a Doppler-free technique by the Stanford group in 1979 [1] opened new possibilities to the use of discharges in high-resolution spectroscopy. Especially the hollow-cathode discharge,where free atoms of refractory elements conveniently can be obtained by sputtering has been given considerable attention lately [2,3,4]. A program for optogalvanic spectroscopy on hollow cathodes was initiated in our group in 1980. Our first experiments in this context using pulsed and CW lasers were performed on a commercial Ne-Ba hollow cathode and a He-Ne laser tube [5]. After additional preliminary experiments on Ne [6] and Cu (unpublished) a comparison between the Doppler-free methods intermodulated optogalvanic spectroscopy (IMOGS) [1], intermodulated fluorescence spectroscopy (IFS) [7] and polarization spectroscopy (PS) [8] was made in connection with studies of isotope shifts in Ne 3s-3p transitions [9]. In the main part of this investigation a hollow cathode with a 6 mm bore, originally designed for classical emission spectroscopy, was used. In order to reduce Stark and pressure broadening a cathode with a larger bore (2.5 cm) designed by Lawler et al. [10] was constructed. For the Ne 3s-3p transitions the new arrangement reduced the homogeneous linewidth by roughly a factor of two and linewidths of 15 MHz have been obtained in subsequent measurements on Mo illustrating the favourable electric field arrangement in this hollow cathode. In Mo the $4d^5 5s\ ^5S_2 - 4d^5 5p\ ^5P_{3,2,1}$ transitions have been studied. Magnetic-dipole and electric quadrupole interaction constants a and b for the upper state have been measured for the two isotopes ^{95}Mo and ^{97}Mo (15.7% and 9.6% abundance) by Meisel [11]. For the a factor of the $^5S_2$ state a value is given in [12,13].

In the next section our set up in hollow-cathode discharge experiments will be described. Section III contains results and discussion. Our results are summarized in a final section.

II - EXPERIMENTAL ARRANGEMENT

In Fig. 1 a schematic diagram of our set up is shown. An argon-ion pumped CR 599-21 single-mode dye laser was employed. The main part of the beam was split into two counterpropagating beams passing the hollow cathode, and the nonlinear interaction in the beam overlap region was detected employing IMOGS, IFS, PS or POLINEX [14]. At A in the figure a polarizer was inserted in the beam in the case of PS and a Pockels cell for modulating the beam polarization between right hand and left hand circular polarization in the case of POLINEX. In the case of PS and POLINEX a $\lambda/4$ plate was also inserted at point B. A squeezed glass plate was used in PS for com-

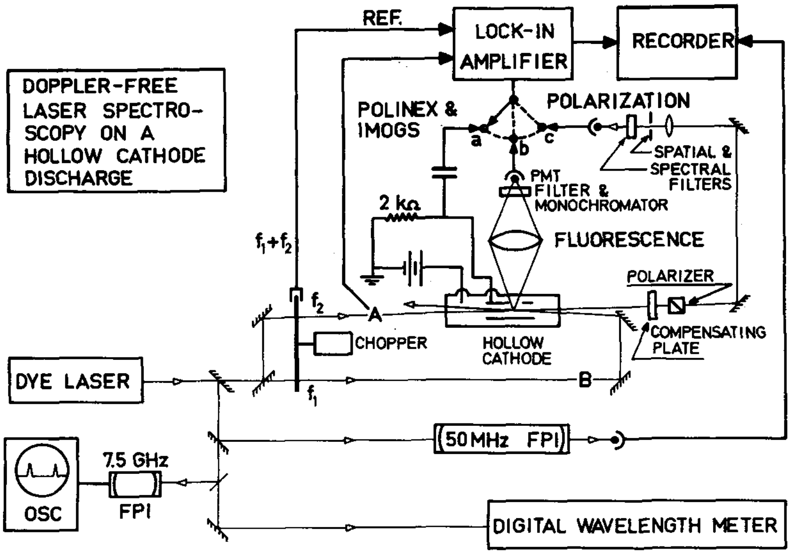


Fig. 1. Experimental set-up. In POLINEX optogalvanic detection was employed.

pensating for birefringence in cell windows. Our matched pair of Glan-Thompson polarizers had an extinction ratio of 10^7. In practical experiments we generally achieved only 10^5 because of the nonuniform birefringence in the cell windows over the area irradiated by the laser beam. By squeezing the cell windows the internal strain in these could possibly have been reduced [8]. Two different choppers were employed; one at 850 Hz for Doppler-limited measurements and one at 750 and 225 Hz for Doppler-free measurements. The choice of chopper frequency was made with some care. The output noise from our CR 599-21 dye laser operating in a single-mode peaks at 127 Hz and 1770 Hz which we attribute to the servo systems. Otherwise the noise of the laser, the hollow cathode and the photomultiplier essentially consisted of white noise up to 50 KHz except for lower harmonics of 50 Hz. Typical data of operation for the hollow cathode in our Mo measurements were a pressure of 0.1 torr and a current of 200 mA. A detailed description of the hollow cathode is given in Ref. [10].

III - RESULTS AND DISCUSSION

In Fig. 2 recordings of the $2p^5 3s\ ^3P_2 - 2p^5 3p\ ^3P_1$ Ne transition at λ=5882 A are shown as obtained with IFS, IMOGS, POLINEX and PS. Table I gives approximative signal/noise ratios obtained with the different methods applied to this Ne transition. Here the discharge current was 45 mA, the pressure 0.1 torr and the lock-in time constant 0.3 s.

Fig. 2.
Experimental curves for the Ne $2p^5 3s\ ^3P_2$ - $2p^5 3p\ ^3P_1$ transition (5882 Å), recorded with

a) IFS, discharge current (I)=150 mA, pressure (p)=0.6 torr, time constant (τ)=3 s.
b) IMOGS, I=45 mA, p=70 mtorr, τ=0.1 s
c) POLINEX, I=45 mA, p=70 mtorr, τ=0.1 s.
d) PS, I=45 mA, p=0.1 torr, τ=0.3 s.

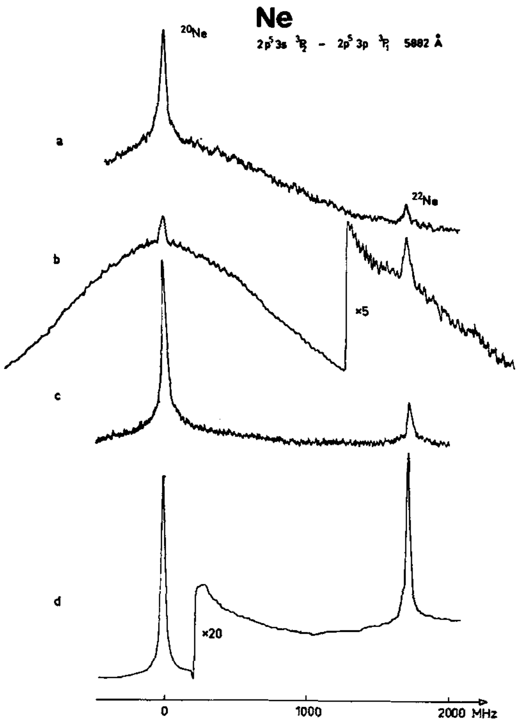


Table I

	Doppler-limited		Doppler-free			
	FS	OGS	IFS	IMOGS	POLINEX	PS
signal/noise	20	100	5	5	30	1500
signal/background	-	-	0.3	0.1	-	-

The term "background" in Table I refers to the Doppler-broadened pedestal due to velocity-changing collisions. These are inherent in the IFS and IMOGS methods and as we shall see, severely limit the usefulness of these two methods.

An interesting aspect concerning spectroscopy on a hollow-cathode discharge is the study of refractory elements. The theory of the hyperfine structure in elements with unfilled d-shell has received considerable attention lately [13,15,16]. The Göteborg group has also recently determined the hyperfine structure in the $4d^5 5p\ ^7P_{4,3,2}$ states in Mo (private communication). We have studied the transitions between the $4d^5 5s\ ^5S_2$ state and the $4d^5 5p\ ^5P_{3,2,1}$ states in Mo. Mo has a relatively low sputtering yield [17]. This means that the currents necessary to produce a fair amount of free Mo atoms in the discharge is rather high. We have used Ne as carrier gas. Other rare gases have a higher sputtering yield [17,18] and might be better suited. In Ref. [19] a unified theory for the sputtering process in a hollow cathode is given. To produce a sufficient amount of Mo atoms in the lowest metastable $4d^5 5s\ ^5S_2$ state the discharge was run at a current of 200 mA and a pressure of 0.1 torr.

Fig. 3 shows two recordings of the $4d^5 5s\ ^5S_2$-$4d^5 5p\ ^5P_3$ transition in Mo at λ=5506 Å. The upper trace is taken with IFS (laser power about 60 mW). In the lower trace taken with PS (laser power <10 mW) transitions corresponding to the five even isotopes

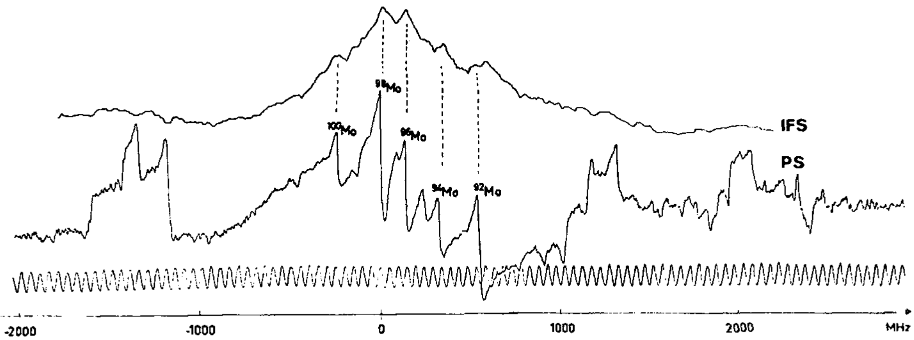


Fig. 3. Experimental curves for the $4d^55s\ ^5S_2 - 4d^55p\ ^5P_3$ transition (5506 Å), recorded with IFS and PS.

and several of the hyperfine transitions in the two odd isotopes with nuclear spin 5/2 can be seen.

Considering Table I, Fig. 2 and Fig. 3 the four alternative techniques will be analyzed.

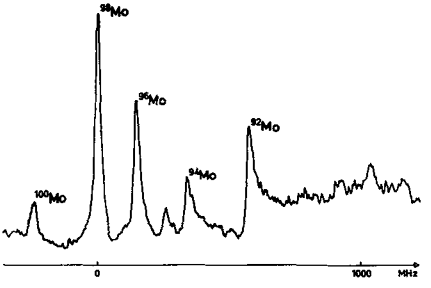
The detectivity in IFS is limited by fluctuations in the broadband pedestal surrounding the Doppler-free peaks. This pedestal emanates from elastic velocity-changing collisions in the lower metastable state. The noise in the pedestal signal is attributed to fluctuations in laser intensity and in the metastable state atom population. Tuning the laser off line decreased the noise level with a factor of four. As also pointed out in Ref. [10], the Doppler-free signal could be enhanced by operating the hollow cathode at higher currents and lower pressure. IFS is more appropriate for weakly absorbing lines [20]. However, in discharges the velocity-changing collisions and fluorescence again decrease the detectivity.

In IMOGS the situation is similar. Here the Doppler-limited pedestal is even more pronounced. This indicates that also other processes than velocity-changing collisions give a Doppler-limited signal on the sum frequency. Our suggestion is that atoms excited by one laser beam indirectly affects the process by which the other laser beam modulates the discharge impedance. This can be explained by e.g. the process suggested in Refs. [21,22], where the ionization is described as a multistep process resulting in a general heating of the plasma. Going to higher currents in order to enhance the Doppler-free signal is less attractive since firstly the relative impedance change due to laser radiation tuned to 3s-3p transitions generally decreases with increasing current [22,23,24] and secondly the demands on the hollow cathode construction for maintaining a quiet discharge increase. The rf discharge might be a good alternative for studying gases, see e.g. [25]. Work in this field is presently going on in our laboratory. The noise in the broadband pedestal was a factor of 15 stronger than the discharge noise when the laser was tuned off line, where it was a factor of 100 above the shot noise limit. The higher currents that had to be used in the Mo measurements caused the discharge noise to rise another factor of five above the shot noise level. This fact in connection with lower sensitivity for the metal atoms than for the metastable Ne atoms [22] caused IMOGS to be unfavourable in this case. In optogalvanic detection the laser beam should have about the same diameter as the cathode to give an optimum in signal/noise. In Doppler-free nonlinear spectroscopy where transitions need to be saturated and in hollow cathodes with large bores sufficient laser power may not readily be obtained for weak lines.

In POLINEX the atomic orientation is probed. If most of the velocity-changing collisions also randomize the atomic orientation, the broad background is eliminated. If the orientation is kept, the background pedestal can still be decreased [26,27] since atoms, which have undergone velocity-changing collisions on the average spend more time before interacting with the second beam. In Ref. [27] this is done by modulating the polarization at a rate where the Doppler-limited and Doppler-free signals come in quadrature and can thus be separated with a lock in amplifier. It would be interesting to investigate if the same procedure could be used to suppress the pedestal in IFS and IMOGS. Further, in Ref. [26] it is demonstrated how the broad component can be partly eliminated by the application of a combination of weak magnetic and rf fields. The detectivity in POLINEX is limited by the discharge noise and could be substantially improved by using a more quiet discharge. However, for studying refractory elements the same problems arise here as for IMOGS.

In our experiments the PS technique clearly gave the best signal/noise ratio. Still several improvements can be made in the experimental set up; e.g. using a more favourable geometry [28], employing a "noise eater" [29], electronically dividing out the laser noise and/or using polarizers as cell windows [30]. The detectivity is limited by birefringence in optical components and laser intensity fluctuations. A drawback with PS is that the signal shapes make the analysis somewhat more difficult. However, for stronger lines heterodyning using a small off-set angle θ may not always be necessary as can be seen in Fig. 4 which shows a recording of the even isotopes in the λ=5506 Å transition of Mo with polarizers crossed and the earth magnetic field compensated. The presence of a small magnetic field otherwise

Fig. 4.
Experimental curve for the $4d^5 5s\ ^5S_2 - 4d^5 5p\ ^5P_3$ transition (5506 Å), recorded with PS. The earth magnetic field has been compensated.



causes a precession of the atomic polarization and in combination with the velocity changing collisions a broad background is obtained [31].

IV - CONCLUSION

In a hollow-cathode discharge we have studied two strong transitions originating in metastable states, one in the carrier gas (Ne) and one in the cathode material (Mo). For these conditions, techniques probing differences in atomic population (IFS, IMOGS) seemed inferior to techniques probing atomic orientation (POLINEX, PS). This conclusion is closely related to the absence of the Doppler-broadened pedestal in the latter techniques as compared to the former ones, for which velocity-changing collisions have a detrimental effect. Lastly we want to point out that other techniques not discussed here such as frequency-modulation spectroscopy [32,33] might prove to be even more efficient in certain cases.

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