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HIGH RESOLUTION INTERMODULATED AND DOUBLE RESONANCE ATOMIC SPECTROSCOPY IN A HOLLOW CATHODE

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Résumé - Les performances d'un tube à décharge à cathode creuse exploité en spectroscopie optogalvanique à haute résolution pour des éléments réfractaires et volatiles sont expliquées. Les structures hyperfines et isotopiques sont résolues.
Les effets de changement de vitesse dû à des collisionnels sont étudiés pour différentes transitions. Des mesures de double résonance optique sans effet Doppler sont présentées pour la première fois avec une détection optogalvanique.

Abstract - The performances of a hollow cathode discharge tube developed for high resolution optogalvanic spectroscopy of both refractory and volatile elements are illustrated. Hyperfine and isotopic structures are resolved. The effect of velocity changing collisions is investigated on different transitions. Doppler-free optical-optical Double Resonance measurements are reported for the first time in optogalvanic detection.

The combination of highly spectral purity Dye lasers with optogalvanic detection scheme has allowed the extention of almost all the nonlinear techniques, typical of absorption spectroscopy, to the whole emission spectrum of an atom. Since the investigations are performed in a discharge, much care is necessary to reduce the perturbations so that developing homogeneously linewidth spectroscopy makes real sense.
In our laboratory we have developed a hollow cathode discharge tube and used it for high resolution spectroscopy of refractory atoms and volatile elements.
Most of the investigations were performed using an active stabilized Coherent 599/21 Dye laser (linewidth about 1 MHz) operating in the rhodamine 6G (or stilbene) tuning range. A second single mode Dye laser was also employed to develope Doppler-free double resonance spectroscopy. Intermodulated spectroscopy allows the resolution of isotopic shifts and hyperfine structures. The comparison of homogeneously broadened signals and Doppler background provides a display of velocity changing collisions. Operation in presence of a magnetic field leads to the resolution of Zeeman effect but also to enhancements of the optogalvanic signal (nonlinear Hanle effect). Versatility and usefulness of the technique are illustrated showing some particular result and starting with the description of the hollow cathode discharge tube.

I - THE HOLLOW CATHODE

The hollow cathode discharge is a well established method to provide gas phase emission spectra of refractory elements. The atoms are ejected from the surface of the cathode as a result of the bombardement by energetic rare - gas ions produced in the discharge. The sputtered atoms which initially posses high kinetic energies rapidly come into thermal equilibrium with the gas as they diffuse from the cathode. The usefulness for atomic spectroscopy has been largely demonstrated also in level crossing and other related experiments which depend upon how the polarization of the fluores-

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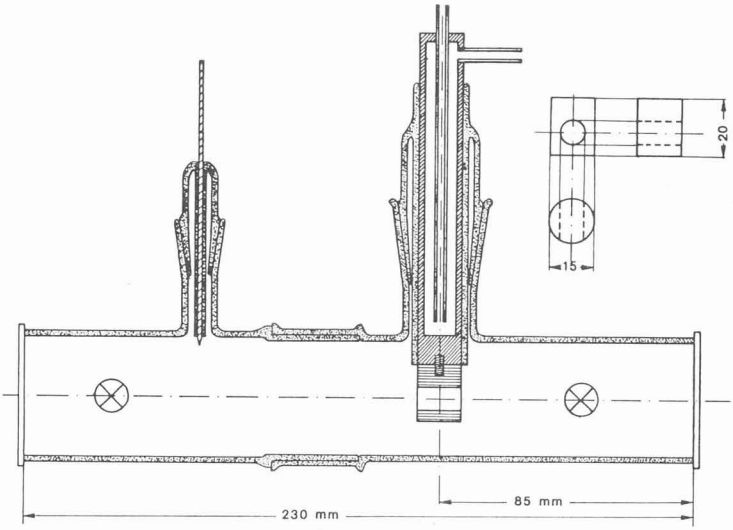

Fig. 1 - Cross - section drawing of the hollow cathode discharge tube.

cence is preserved in the presence of the rare gas and the discharge /1/.
The development of highly sensitive methods of Doppler - free laser spectroscopy, also combined with the optogalvanic detection scheme, has made possible the study of atomic levels even sparsely populated after sputtering in the gas discharge. As a consequence also the homogeneous linewidths which can be obtained in a hollow cathode configuration become important. In fact since a discharge does not provide an ideal environment for high resolution laser spectroscopy (there are perturbations due to neutral and charged particle collisions and due to electric fields), observed homogeneous linewidths are an indication of the severity of such perturbations. Narrow homogeneous widths can be achieved using large cathode diameters which allow operation at lower gas pressure, reduced space-charge density, and electric fields. The increase in the cathode diameter causes also a decrease in the current density and in refractory atoms density. As a consequence the design of a hollow cathode for high resolution optogalvanic spectroscopy must take in account the request for an easy demountability and change in the geometrical characteristics in order to optimize the operation for the given element and transition. Fig. 1 shows the cross section of the hollow cathode discharge tube constructed in our laboratory. The vacuum tightness is assured by commercial pyrex conical connections. They are demountable for sample, cathode, anode changes and provide high vacuum seal when remounted. The anode is a 20 mm - diameter tungsten pin or a 50 mm - diameter aluminium rod. Once optimized all the parameters, no significant differences have been observed between the two except that clean discharge operation seemed easier to be obtained with aluminium. As for the cathode sample, in the simplest case (Copper) it was constituted of a Cu tube with internal diameter up to 12 mm. Different solutions have been adopted for other materials. For instance in case of Ca, metallic calcium have been melted into a stain less steel cylinder and then a hole has been drilled. In case of U, a rectangular

thin piece has been rolled into a copper cylinder. Water cooling is provided for the cathode holder, shown in more detail in the upper - right part of the figure. All the metallic parts in the discharge tube but the final portion of the cathode holder and the point of the anode are shelted by glass tubes. It is essential that all the windows be located far enough from the cathode to prevent a sputtered metal coating. All the device is relatively simple to construct, and uses a small amount of sample material (for instance in case of uranium about 0.8 grams were used). Neon and argon were used to operate the discharge. Clean operation could be obtained for gas pressures down to .4 Torr and for current intensities from 1 to 80 mA.

II - INTERMODULATED SPECTROSCOPY

Thanks to the good optical accessibility of the internal portion of the cathode where the discharge is localized, laser-sample interaction geometries are easy obtained which allow subdoppler recordins of optogalvanic signals. General performances of the system can be deduced from the measurements reported in ref./2/. The Cu transition at 578.2 nm was investigated. The measurement using a unidirectional laser beam displaied a Doppler width definitively lower than the previously reported for the same transition in the literature.It could be computed a kinetic temperature T= 512 ^{o}K to be compared with T= 1025 ^{o}K and T= 1457 ^{o}K respectively estimated from /3/ and /4/. The lower kinetic temperature in our apparatus is caused by the water cooling and the lower current intensity (50 mA in the recording of /2/). The Doppler-free intermodulated spectrum was also recorded. The signal to noise ratio was almost comparable to other spectra reported /3,4/using hollow cathodes but detecting transmitted or fluorescent light. It should be added that the better signal to noise ratio obtained with intermodulated fluorescence spectroscopy /5/ requires in general the elimination of

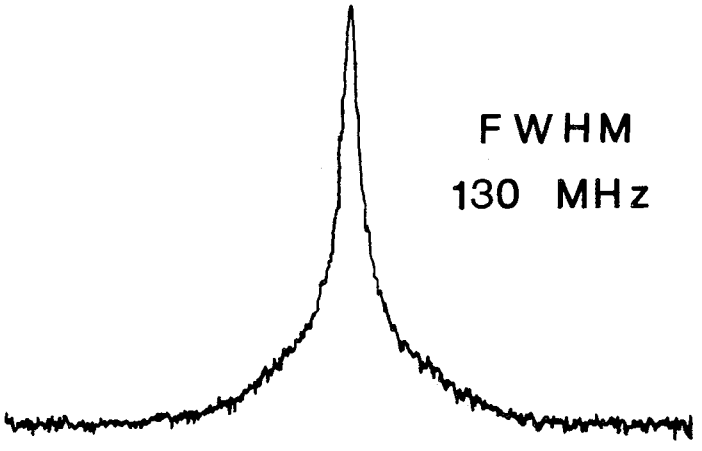

Fig. 2 - Doppler-free intermodulated optogalvanic spectroscopy recording of Calcium $^3P_1$-$^3S_1$ transition at 612.2 nm. Argon was used with a pressure of 1.1 Torr. I=50 mA.

the noise of the scattered laser light. This is usually obtained by detecting the fluorescence on associated transitions in different spectral ranges. When this possibility is not available, the optogalvanic detection is the only one available with signal to noise ratio good enough to perform high resolution spectroscopy. Calcium transitions connecting the metastable $^3P_{0,1,2}$ levels to the $^3S_1$ provide an experimental situation of absence of associated fluorescence. Nevertheless (Fig. 2) an intermodula-

ted optogalvanic signal could be obtained with good signal to noise ratio. That was preserved even when the discharge was operated in presence of a static magnetic field and made possible subdoppler Zeeman optogalvanic spectroscopy measurements /6/. In the recording of Fig. 2 the homogeneously broadened signal (Lorentzian) is well detected over a small background due to velocity changig collisions /7/. It is worth noting the not particularly pronounced contribution of velocity changing collisions in spite of the long lifetime of the metastable lower level. On the other hand the small effect of the collisions on the Ca metastable triplet in a discharge have been recently demonstrated directly investigating the broadening of the levels by means of optogalvanic nonlinear Hanle effect /8/. This technique has allowed to observe with the apparatus described in the present paper subhomogeneous linewidth structures, corresponding to a level FWHM of 8 MHz /8/ and to be published.

III - VELOCITY CHANGING COLLISIONS - Neon

The results of previous paragraph for the metastable state of Ca were possible thanks to the small collisional cross sections. It should be noted that in general the importance of velocity changing collisions is difficult to be stated a priori simply on the basis of atomic mass and levels lifetimes. For instance,differently from Ca, in case of Cu the effect was actually enhanced /2/ by the relatively long lifetime of the metastable lower level ($^2D_{3/2}$). The importance of the lower level lifetime can be evi-

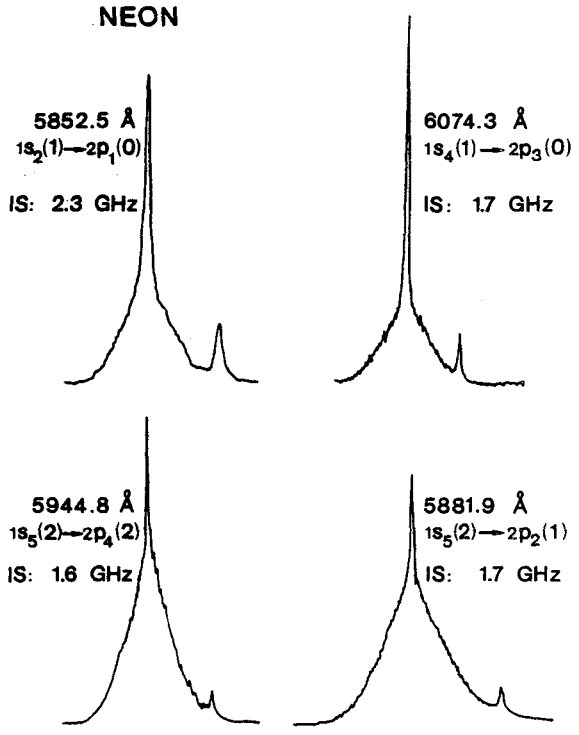

Fig. 3 - Doppler-free intermodulated recordings of natural Neon transitions involving (lower two) or not (upper two) metastable levels. The values of the 20 - 22 isotopic shifts have also been reported.

denced by performing measurements on the same element, with similar pressure and discharge conditions, but choosing transitions involving levels with different lifetimes.Typical results are shown in Fig. 3 for Ne. The velocity changing collisions

background is significantly more pronounced for transitions starting from metastable levels. The values of gas pressure were .55, 1.2, .65, and .60 Torr going from left to right and up to down in the Figure. It is worth noting the better doppler-free doppler broadened signals ratio for the transition at 607.4 nm in spite of a pressure value which was about twice the one used for the transitions starting from metastable levels.

IV - URANIUM

The atomic mass by itself seems to be less important in affecting velocity changing collisions. That will be illustrated using the heavy Uranium atom. U was sputtered in the Ne gas discharge as described in first paragraph. A sample with 235 isotope enriched to 6% was used for the measurements shown in Fig. 4. Doppler broadened record-

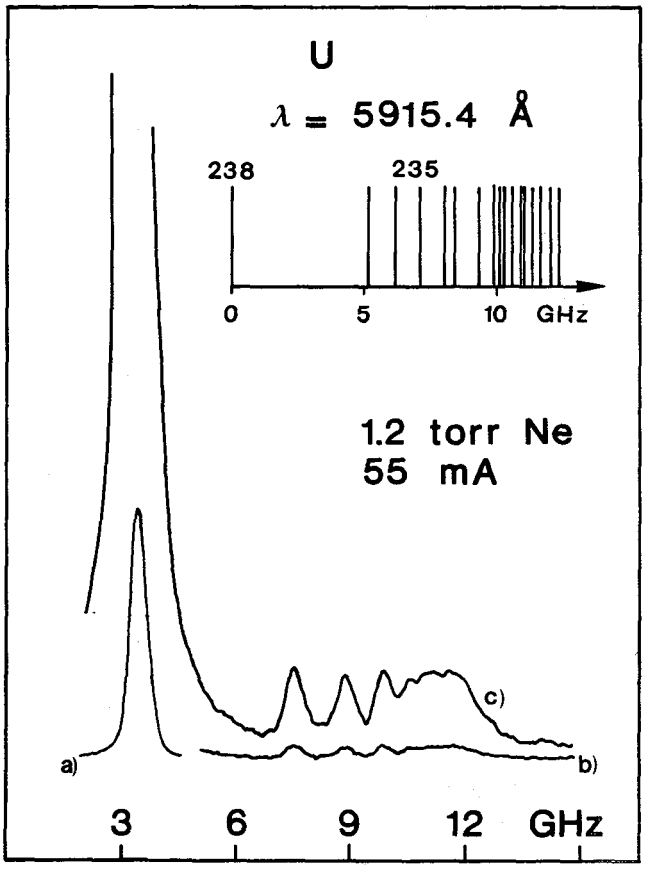

Fig. 4 - Doppler limited optogalvanic recordings with a U sample enriched to 6% of 235 isotope.The total metal amount used in the hollow cathode was about .8 grams. a) X1; b) X3; c) X18.

ings obtained with about 50 mW of laser power are reported. From the Doppler widths it is obtained a kinetic temperature of 760°K, a little higher than for Cu but still low enough to allow a first partial resolution of the hyperfine structure. The observed envelope of the shift and hyperfine components of the 235 isotope is consistent with data available in literature and schematically shown in the insert of Fig. 4. A complete resolution of the hyperfine structure could be obtained by intermodulated optogalvanic spectroscopy /to be published/. In this case the signal to noise ratio and resolution were comparable to those of previous photoionization measurements on an atomic beam /10/. On the contrary the relative intensity of the individual compo-

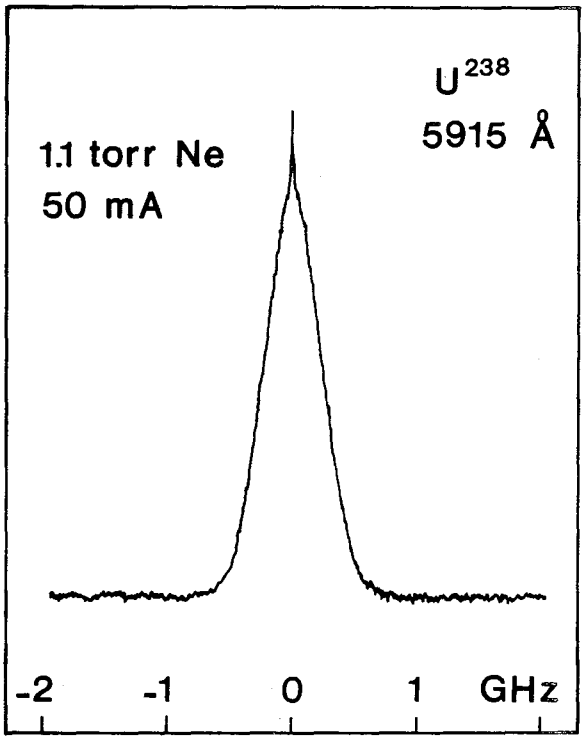

Fig. 5 - Doppler-free recording of the 591.5 nm transition in ^{238}U.

nents was not in agreement with the theoretical relative strengths, confirming that caution must be used in the quantitative analysis of the intensity of lines detected by means of optogalvanic spectroscopy.

An intermodulated optogalvanic recording of the isotope 238 is shown in Fig.5. The laser beam (70 mW) was splitted into two counterpropagating beams of nearly the same intensity. The high Doppler background confirms the consideration of previous paragraph that that the atomic mass by itself is not determinant in reducing the effect of velocity changing collisions (the transition starts from the ground level).

Of course one of the advantages of hollow cathode optogalvanic technique is that the investigations can be extended to levels of refractory elements other than the ground one.For instance in Fig. 6 it is shown a compared quantitative analysis of optogalvanic signals for two U transitions starting from the ground state (439.4 nm) and from the metastable one at 620 cm^{-1} (435.6 nm). Measurements were performed at constant laser intensity (about 150 mW) as a function of the discharge current. Signals increase almost linearly up to a constant value. This value is reached at lower current intensities for the ground state transition, but the final signal intensities are almost comparable.

Another effect which can be used to enhance an optogalvanic signal is the nonlinear Hanle effect /11/ which is caused by the different saturation of an atomic transition when an external magnetic field removes the M sublevels degeneracy. Increases of the optogalvanic signal higher than 10% are for instance shown in Fig. 7 for the U transition at 591.5 nm. In that case the hollow cathode discharge was operated in presence of a magnetic field parallel to the current and to the laser beam and orthogonal to the linear polarization of the latter. Dashed curve refer to a possible Fit to a Lorentzian for the low magnetic field points, which can lead to an evaluation of the saturation of the transition /8,11/.

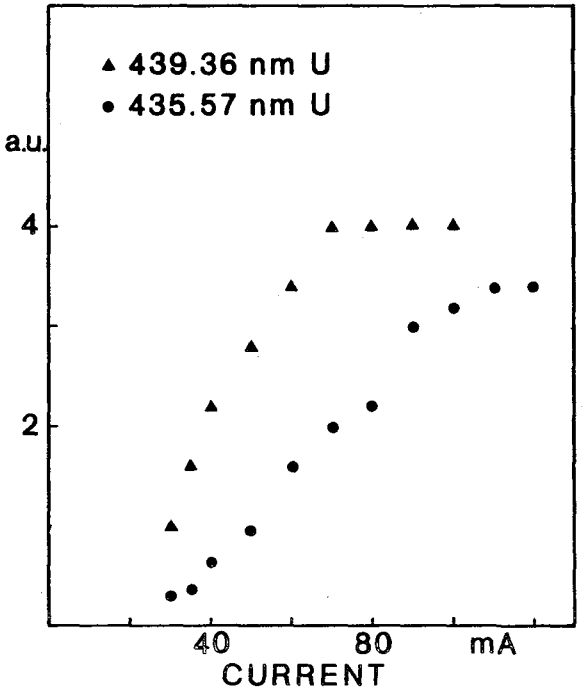

Fig. 6 - Comparison of different transitions signals from U sputtered in the hollow cathode.

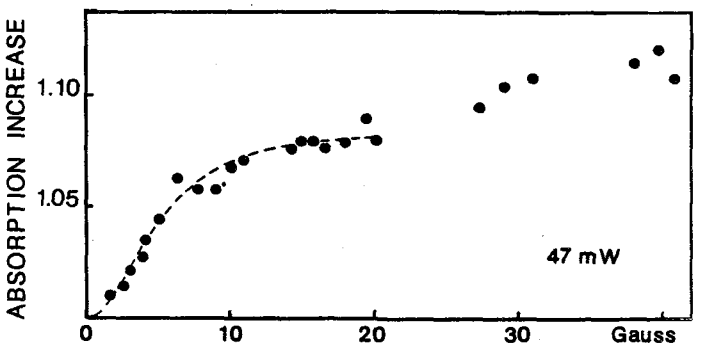

Fig. 7 - Enhancement of the o.g. signal from U at 591.5 nm caused by the nonlinear Hanle effect.

V - OPTOGALVANIC DOUBLE RESONANCE

Optical - optical double resonance optogalvanic detection had been proposed /12/ as a method for state selective spectroscopy in plasmas. The efficiency of the technique in getting simpler complex molecular spectra was recently demonstrated /13/. The technique can be used to determine a common intermediate energy level but can also lead to interesting information concerning energy transfer processes in atoms or molecules when the two transitions are coupled via collisions. For instance this possibility was experimentally evidenced in /14/ . It is worth noting that both in /13/ and /14/ Doppler free signals were not reported. On the contrary that is an important feature to be investigated, in particular for a study of the mechanism of energy transfer. For instance the recent direct observation of heating of the discharge obtained combining

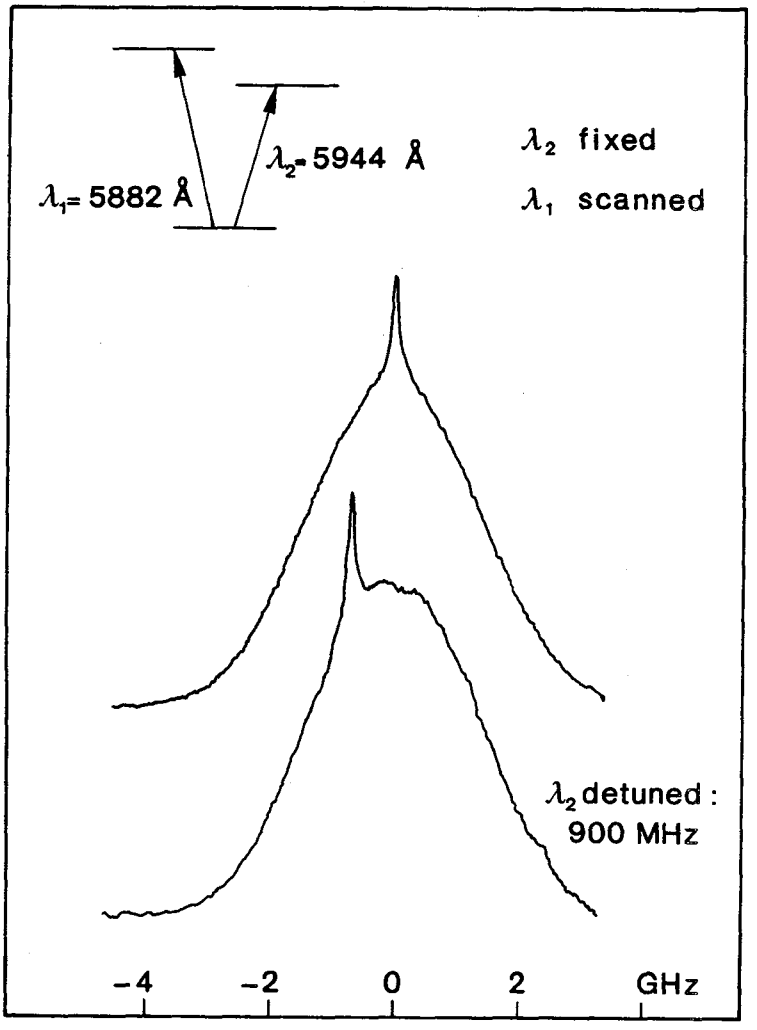

Fig. 8 - Experimental demonstration of optical - optical double resonance Doppler-free optogalvanic spectroscopy in Ne. The two laser beams intensity was about 100 mW and the gas pressure 1 Torr.

optoacoustic and optogalvanic detections /15/ opens the question if energy transfers are caused by simple atom-atom collisions or if there is an important intermediate role plaied by the electrons. Of course the two effects can be discriminated since the first can lead to Doppler-free observations.

We report here the experimental observation of Doppler-free optogalvanic double resonance spectroscopy. Two counter-propagating single mode laser beams were fed into the hollow cathode discharge, each tuned on one Ne transition and amplitude modulated at a different acoustic frequency. The optogalvanic signal was detected at a frequency sum of the two. As shown in Fig 8, the wavelength of one of the lasers was fixed and the signal was recorded by scanning the wavelength of the other. The homogeneously

broadened signal is well evident over the large background caused by velocity changing collisions. Note that the common level shared by the two transitions is metastable, as shown also in Fig. 3. Different atomic velocity groups can be selected by changing the wavelength of the fixed laser within the Doppler profile. We think that the good signal to noise ratio observed opens the possibility of investigating velocity changing or not collisions in energy transfers in plasmas.

In conclusion the results illustrated in the present paper show that high resolution laser spectroscopy of refractory and volatile atoms can be successfully performed using the simple hollow cathode developed.

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