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Optogalvanic detection of barium high-lying levels with a two-step pulsed laser excitation

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Résumé. — Les niveaux très élevés du baryum neutre ont été explorés en utilisant une excitation laser pulsée à deux étages avec une détection optogalvanique. On excite les atomes à partir des niveaux métastables $5d\ 6s\ ^3D_{1,2,3}$, peuplés par une décharge dans une vapeur du type *heat-pipe*. On atteint les configurations $5dnl$ paires par l'intermédiaire des niveaux de la configuration $5d\ 6p$. Des niveaux ayant des valeurs de J comprises entre 0 et 5 peuvent ainsi être observés. On présente les résultats préliminaires.

Abstract. — Highly excited levels of neutral barium have been investigated by using a two-step pulsed laser excitation combined with an optogalvanic detection. The excitation process starts from the $5d\ 6s\ ^3D_{1,2,3}$ metastable levels that are populated in a heat-pipe discharge. It leads to the even parity $5dnl$ configurations via the $5d\ 6p$ intermediate levels. Levels with J values ranging from 0 to 5 can be observed. Preliminary results are given.

Until now observations of highly excited levels in multiphoton or multistep spectroscopy have been generally limited to processes starting from the ground state. In a two-photon or a two-step process, the investigation is limited to upper levels with J , $J \pm 1$, $J \pm 2$ values, where J is the angular momentum quantum number for the ground state. In barium the ground level has a $J = 0$ value, so with a two-photon process the investigation is limited to upper levels with $J = 0$ or 2, and with a two-step process to upper levels with $J = 0$, 1 or 2. In order to extend the J range, processes using more than two photons or two steps are needed. But the experimental arrangement becomes more complex, and particularly the analysis of the recordings obtained is not so obvious owing to the increasing number of multistep processes that occur simultaneously.

Under these conditions, further investigations are needed with optical transitions from excited levels populated by a non optical process. A glow discharge provides an easy way to populate several excited levels. In this case, transitions from these levels to various upper levels can occur, thus we can obtain a large range of J values.

Resonant optical transitions in a discharge can be detected by means of the optogalvanic effect. It is well known that irradiation of a gas discharge with a laser, tuned to an atomic transition occurring within the discharge, produces impedance changes

that can be detected as changes in the potential across the discharge tube [1, 2, 3, 4]. Recent applications of this effect have been reported; for most of them, modulated c.w. laser excitation is used.

In our experiment with barium, we carry out a two-step excitation process, starting from metastable levels, with a pulsed dye laser excitation, and we detect the transient optogalvanic signal in a heat-pipe discharge cell. With these techniques, we plan to extend our previous observations with barium [5] to other J values and configurations.

The experimental arrangement is shown in figure 1. A Moletron UV-1 000 nitrogen pulsed laser is used

Fig. 1. — Experimental set-up for optogalvanic spectroscopy.

to pump two similar dye lasers, with a 15 Hz repetition rate. The pulse duration is about 7 ns and the laser linewidth is 0.3 cm^{-1} . The two dye laser beams are passed through a cylindrical heat-pipe discharge tube as previously described by P. Camus [6] (for convenience the two beams counter propagate axially through the tube).

The discharge tube has an 8 mm internal diameter and an active length of nearly 180 mm. Helium is used as a buffer gas. The barium vapour is contained in a stainless steel pipe, 155 mm long, in a vapour pressure range of 2.3-2.7 T (corresponding to temperatures between 905 and 935 °C). Thus stable discharges have been obtained with a constant potential of 1.45 kV applied across the tube and a 51 k Ω ballast resistor. Under these conditions the tube operates at 20 mA-400 V.

The a.c. component of the potential across the tube is coupled to an oscilloscope and simultaneously to an electronic recording device, through a 6 800 pF blocking capacitor and a band-pass active filter. The filter has a 110 kHz central frequency and a 90 kHz bandwidth. Its gain for the optogalvanic signals, we usually observed, is about 17.

The first dye laser is tuned to the first step transition. The wavenumber of the second varies in order to explore highly excited levels in the discrete and self-ionized regions. (For calibration, a small part of the dye laser beam is passed through a Fabry-Perot interferometer.)

When interacting with the pulsed dye laser tuned to an atomic transition, the perturbed discharge returns to its previous steady state by a transient optogalvanic response as shown in figure 2. We observe such signals for transitions occurring either in barium or in helium. The shape of the optogalvanic signal probably arises from the characteristics of the electric circuit on the one hand, and from induced changes in the discharge internal impedance on the

other hand, corresponding to rearrangements between the populations of excited levels. This oscillating shape varies with the interacting element, helium or barium, and slightly with the transition involved.

Peak amplitude detection has been chosen : the signal is taken as the potential at the first extremum (see for example figure 2, which gives a negative signal corresponding to a drop in the potential across the tube). It seems to us that this signal reproduces with sufficient accuracy the population changes induced, that is to say, that, in this way, line heights are nearly proportional to the laser peak power and oscillator strength of the transition involved.

The instrumental linewidth is determined from the dye laser linewidth, approximately 0.3 cm^{-1} , which is wider than the Doppler width (0.06 cm^{-1}). The sensitivity of the detection is limited by discharge noise to 3.5 mV. In practice, our electronic device limits the input signal to 1 V, so the dynamics range of detection is about 300. But this is not a fundamental limitation, the intrinsic optogalvanic signal dynamics range can probably reach 10^4 - 10^5 . A well adapted electronic system will be used in our future experiments.

In the discharge, the metastable levels $6s 5d \text{ } ^3\text{D}_{1,2,3}$ are well populated, compared to other excited levels, and give rise to strong optogalvanic signals when optical resonant transitions involve one of them. In order to observe highly excited levels of $5dnl$ configurations, we perform a two-step excitation process starting from the $6s 5d \text{ } ^3\text{D}$ metastable levels via intermediate levels of the $5d 6p$ configuration (see figure 3).

Since we can choose intermediate levels with J values ranging from 0 to 4, even levels belonging to $5dns$ and $5dnd$ can be observed with J values equal to 0, 1, 2, 3, 4 or 5 ; that is to say, all levels of $5dns$ and $5dnd$ configurations are liable to observation considering the J selection rule alone.

Fig. 2. — Optogalvanic signal occurring for a barium resonance. The calibrations are : vertical 500 mV/division, horizontal 5 μ s/division.

Fig. 3. — Barium levels diagram.

Fig. 4. — Two-step optogalvanic spectrum of barium. *a*) Fabry-Perot fringes, $\Delta\sigma = 1.314\,67(8)\text{ cm}^{-1}$; *b*) Spectrum of Ba with the $5d\,6p\,^3P_2^0$ intermediate level in the range $6\,281\text{--}6\,315\text{ \AA}$.

Moreover, we can discriminate the two-step excitation process from any other process by comparing two spectra : one recorded without first step excitation, the other recorded with two-step excitation. The lines that appear alone or that are strongly enhanced in the two-step spectra correspond to transitions from $5d\,6p$ to $5dnl$, as shown in figure 6.

In this way, we have observed several highly excited levels in the energy region of the $5d\,7d$ configuration, which is close to the $6s\,^2S_{1/2}$ first limit

$$(\sigma = 42\,035.05\text{ cm}^{-1}),$$

and in the energy range just below the $5d\,^2D_{3/2,5/2}$ limits

$$(\sigma(^2D_{3/2}) = 46\,908.9\text{ cm}^{-1},$$

and

$$\sigma(^2D_{5/2}) = 47\,709.9\text{ cm}^{-1}).$$

In the discrete spectrum close to the first limit, the $5d\,7d$ levels appear as perturbators of the $6snl$ series. Therefore, a few members of the $6snl$ series are observed only in the vicinity of the perturbator; transitions from $5d\,6p$ to $6snl$ are not observed elsewhere owing to the excitation of two electrons. The most important process is configuration mixing ($5d\,7d + 6snl$), the highest line intensities involve $6snl$ levels. Figure 4b shows the spectrum obtained in the region of the $5d\,7d\,^1D_2$ level with the perturbed members of the $6snd\,^1,^3D_2$ series, that we have previously observed [5]. In this case, the second step transition occurs from the $5d\,6p\,^3P_2^0$ level. The increase and decrease in line intensity can be seen on both sides of the perturbator; the maximum intensity is observed for $n = 26$ and 27 members. Moreover, this observation closely corresponds to the admixture of the $5d_{5/2}\,nd_{3/2}$ channel given in the MQDT study of the $J = 2$ levels [7].

This phenomenon is quite useful to identify at the same time as some recently observed perturbators and perturbed series members. The observation of many spectra with different intermediate $5d\,6p$ levels and comparisons between them, give the energy of the levels, using the Ritz's principle, and their J value according to the selection rule.

Figure 5b shows a small part of the spectrum obtained with $5d\,6p\,^3P_1^0$ as the intermediate level, and several lines observed leading to the determination of unknown energy levels. The strongest lines observed in the vicinity of the $5d\,7d\,J = 1$ level correspond to transitions from $5d\,6p\,^3P_1^0$ to $6s\,19s\,^3S_1$, and $6s\,20s\,^3S_1$, and to one transition from $5d\,6p\,^3P_1^0$ to $6s\,18d\,^3D_1$. In the table, observed wavenumbers and intensities are given for these transitions observed in three different spectra, obtained with $5d\,6p\,^3P_0^0$, $^3P_1^0$ and $^3P_2^0$ intermediate levels. The observations of several upper levels belonging to the $6sns\,^3S_1$ and $6snd\,^3D_1$ series have been extended to n values between 19 and 41, and between 17 and 36 respectively.

Fig. 5. — Two-step optogalvanic spectrum of barium. *a*) Fabry-Perot fringes, $\Delta\sigma = 1.314\,67(8)\text{ cm}^{-1}$; *b*) Spectrum of Ba with $5d\,6p\,^3P_1^0$ intermediate level in the range $6\,294\text{--}6\,320\text{ \AA}$.

Table I. — *New observed transitions in barium occurring in a two-step excitation process via three intermediate levels of the 5d 6p configuration.*

		Intermediate odd levels used		
		5d 6p ³ P ₀ ^o	5d 6p ³ P ₁ ^o	5d 6p ³ P ₂ ^o (*)
New even levels observed and their average energies		25 642.10 cm ⁻¹	25 704.08 cm ⁻¹	25 956.49 cm ⁻¹
6s 19s ³ S ₁	41 527.08 cm ⁻¹	15 885.10 S	15 822.96 VS	15 570.51 w
6s 18d ³ D ₁	41 558.97 cm ⁻¹	15 916.87 VS	15 854.96 VVS	15 602.42 m
5d 7d J = 1	41 570.24 cm ⁻¹	15 928.23 VVS	15 866.05 VVS	15 613.76 m
6s 20s ³ S ₁	41 592.74 cm ⁻¹	15 950.56 VS	15 888.69 VVS	15 636.37 w

w : weak
m : medium
S : strong
V : very

Intensities of the lines.

(*) The 5d 6p ³P₂^o intermediate level has been populated by dye laser fluorescence.

Another perturbator level 5d 7d J = 1 is obtained [$\sigma = 41\,930.87\text{ cm}^{-1}$] located between 6s 36s ³S₁ and 6s 37s ³S₁ and also between 6s 35d ³D₁ and 6s 36d ³D₁.

Moreover, two 5d 7d J = 3 levels have been recently observed and identified :

$$\sigma_1(5d\,7d\,J=3) = 41\,459.23\text{ cm}^{-1}$$
$$\sigma_2(5d\,7d\,J=3) = 40\,972.82\text{ cm}^{-1}.$$

They perturb the 6snd ³D₃ series, members of which have been observed, for the first time, for n ranging from 11 to 17. Many other highly excited levels have also been observed in this region ; some of them

are identified, but the complete analysis is in progress.

In the self-ionized region we have undertaken the observation of the 5dns and 5dnd Rydberg series, close to the 5d ²D_{3/2} limit. Figure 6 shows part of the observed spectrum, where the second step transition occurs from the 5d 6p ³P₁^o intermediate level. The comparison of two spectra, one with, and the other without the first step, shows clearly that several lines appear when the first step is applied. The observation of the self-ionized region, from 6s ²S_{1/2} up to 5d ²D_{3/2, 5/2} is currently under way.

Our observations are complementary to those recently made by Wynne *et al.* [8] with a two-step experiment using a thermoionic diode, for transitions

Fig. 6. — Optogalvanic spectra of barium in the range 4 732-4 755 Å. a) Fabry-Perot fringes ; b) Spectrum of Ba without first step excitation ; c) Spectrum of Ba with first step transition 5d 6s ³D₂-5d 6p ³P₁^o.

from the $6s^2\ ^1S_0$ ground state via the $5d\ 6p\ ^1P_1^0$ intermediate level. The investigation of highly excited states in an excited discharge with optogalvanic detection is obviously a very powerful and fruitful method. For example the observation of $J = 5$ levels in a conventional multistep excitation process from the ground state would need no less than five steps. Beside those with two dye lasers, any level of the $5dns$ and $5dnd$ configurations in barium can be

observed using optogalvanic detection spectroscopy provided the oscillator strengths are not too small. However, with the high peak power of the dye laser and the sensitivity of the optogalvanic detection, very weak transitions can be observed.

We plan to obtain a complete description of the $5dns$ and $5dnd$ configurations; for this purpose, we are extending our experimental observations, and the analysis of the spectra is in progress.

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