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RELAXATION OF THE $4D_{3/2}$ AND $4D_{5/2}$ LEVELS OF SODIUM ATOM PERTURBED BY NOBLE GASES

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Résumé. — L'atome de sodium est excité sélectivement dans le niveau $4D_{3/2}$ ou le niveau $4D_{5/2}$ par la méthode d'absorption à deux photons sans élargissement Doppler. Nous analysons la fluorescence en intensité et en polarisation en présence de gaz rares. Nous en déduisons les sections efficaces correspondant au transfert de population entre $4D_{3/2}$ et $4D_{5/2}$ et entre $4D$ et $4F$, la relaxation de l'alignement dans chacun des sous-niveaux ainsi que le transfert d'alignement. Nous avons d'ailleurs observé un transfert d'alignement dans le cas de collisions contre l'hélium et le néon. Ces résultats sont comparés à un calcul semi-classique utilisant les potentiels de Pascale et Vandeplanque.

Abstract. — The sodium atom is excited selectively in the $4D_{3/2}$ or $4D_{5/2}$ level using Doppler-free two-photon absorption. We analyse the fluorescence in intensity and polarization in presence of noble gas atoms. We measure the cross-sections corresponding to the transfer of population between $4D_{3/2}$ and $4D_{5/2}$ and between $4D$ and $4F$, the relaxation of alignment in both fine sublevels and the transfer of alignment. In particular, we have observed a transfer of alignment in the case of helium and neon perturbers. These results are compared with semi-classical calculations performed using the potentials of Pascale and Vandeplanque.

Doppler-free two-photon spectroscopy [1] has been shown to be a very powerful method for studying collision processes. In two previous papers [2, 3] we have reported precise measurements of the collisional shift and broadening of sodium two-photon lines perturbed by noble gases. It is easy to perform such measurements because the two-photon line is free of Doppler broadening and the linewidth is very sensitive to small amounts of a foreign gas. Another application of Doppler-free two-photon spectroscopy is to excite one sublevel inside the Doppler width and to observe the relaxation of this sublevel (transfer to other levels [4], depolarization...). We describe hereafter several experiments of this type performed on the $4D_{3/2}$ and $4D_{5/2}$ levels of sodium atom perturbed by noble gases. The experimental results are compared to theoretical predictions. We do not intend to present here in detail all the results obtained. We just want to summarize the essential features of our investigation. A more complete paper is in preparation.

1. Description of the experiment. — The experiment is performed on the $3S$ - $4D$ two-photon transition in sodium [5, 6]. The $4D$ level is split into two sublevels $4D_{3/2}$ and $4D_{5/2}$ due to the spin orbit coupling.

FIG. 1. — Energy levels of sodium which are involved in the present experiment.

These two sublevels are separated by 1.03 GHz (the Doppler width on the two-photon line is about 3 GHz). The Doppler-free two-photon excitation is used in order to selectively populate either the $4D_{5/2}$

or the $4D_{3/2}$ level. Furthermore, the $4D_{5/2}-3P_{1/2}$ line is forbidden in the electric dipole approximation, it follows that the $4D_{5/2}$ level decays toward the $3P_{3/2}$ (5 688 Å line, see figure 1). In the case of the $4D_{3/2}$ level, the intensity of the $4D_{3/2}-3P_{1/2}$ line (5 682 Å) is five times larger than the intensity of the $4D_{3/2}-3P_{3/2}$ line (5 688 Å). We conclude that it is possible by analysing the fluorescent light with a monochromator to determine the transfer between the two sublevels $4D_{3/2}$ and $4D_{5/2}$.

In fact, the fluorescent light is analysed in intensity and in polarization. We can thus measure the relaxation of the polarization within each sublevel, and also the transfer of polarization between the two sublevels.

In our experiment, the two counter propagating beams are circularly polarized [6]. The fluorescence light is detected in a direction perpendicular to the direction of propagation of the exciting light. The polarization of the analyser located at the entrance of the monochromator is linear and is either parallel to the direction of propagation of the exciting light (I_π) or perpendicular to it (I_σ). In order to calibrate the transmission of the optical detection for the two polarizations, we have used the 3S-5S two-photon transition in sodium [7, 8]. For this case the two-photon operator is scalar and the fluorescent light is not polarized [9].

In the present experiment the temperature of the sodium cell is 270 °C. We present the results concerning the collisional transfer of population between $4D_{3/2}$ and $4D_{5/2}$, and between the 4D and 4F levels and the effect of collisions on the polarization of the fluorescent light (depolarization and transfer).

2. Collisional-transfer of population between $4D_{3/2}$ and $4D_{5/2}$. — The experimental method can be understood from figure 2 : the atom is excited in the $4D_{5/2}$ level, and by scanning the wavelength of the monochromator (the frequency of the laser being fixed) one obtains a fluorescence spectrum which depends on the pressure of the foreign gas. If there is

FIG. 2. — Fluorescence spectrum obtained when the frequency of the laser is locked on the $3S_{1/2}-4D_{5/2}$ two-photon transition in sodium for different pressures of neon in the cell. The exciting frequency is fixed and the wavelength of the monochromator at the detection is scanned.

no noble gas, there is only one line in the fluorescence spectrum ($4D_{5/2}-3P_{3/2}$). In presence of a noble gas there is a transfer induced by collision between $4D_{5/2}$ and $4D_{3/2}$ and the line at 5 682 Å corresponding to the $4D_{3/2}-3P_{1/2}$ transition appears in the spectrum. The intensity of the collision induced line increases with the pressure of the noble gas.

In fact, in order to analyse with precision the intensities, one has also to take into account the possibility of quenching. By studying the energy diagram of sodium, it appears that the only level which can be connected to 4D in a collision with a noble gas atom is the 4F level (which is only 40 cm^{-1} away, see figure 1). It follows that the rate equations are the following : ($N_{5/2}$, $N_{3/2}$ and N_{4F} are the populations of the $4D_{5/2}$, $4D_{3/2}$ and 4F levels)

$$\frac{d}{dt} \begin{bmatrix} N_{5/2} \\ N_{3/2} \\ N_{4F} \end{bmatrix} = \begin{bmatrix} -\frac{1}{\tau} - \left(G_0 + \sqrt{\frac{2}{3}} \frac{3/2}{5/2} \varphi^0 \right) & \sqrt{\frac{2}{3}} \frac{3/2}{5/2} \varphi^0 & 0 \\ \sqrt{\frac{2}{3}} \frac{3/2}{5/2} \varphi^0 & -\frac{1}{\tau} - \left(G_0 + \sqrt{\frac{3}{2}} \frac{3/2}{5/2} \varphi^0 \right) & 0 \\ G_0 & 0 & -\frac{1}{\tau'} - \frac{5}{7} \Gamma_0 \end{bmatrix} \begin{bmatrix} N_{5/2} \\ N_{3/2} \\ N_{4F} \end{bmatrix} + \begin{bmatrix} {}^{5/2}\lambda_0 \\ 0 \\ 0 \end{bmatrix} \quad (1)$$

${}^{5/2}\lambda_0$ is the pumping rate when the frequency of the laser is tuned on the $3S_{1/2}-4D_{5/2}$ transition. τ and τ' are the lifetimes of the 4D and 4F levels. $\frac{3/2}{5/2}\varphi^0$ corresponds to the collision induced transfer of population between $4D_{3/2}$ and $4D_{5/2}$. The $\frac{3/2}{5/2}\varphi^0$ coefficient

is *symmetrical* ($\frac{3/2}{5/2}\varphi^0 = \frac{5/2}{3/2}\varphi^0$) ⁽¹⁾. G_0 and

⁽¹⁾ These coefficients appear in a natural way by using a decomposition of the density matrix on an irreducible tensorial set basis (Refs. [10] and [11]).

Γ_0 correspond to the transfer of the population between 4D and 4F, they are related by the relation :

$$G_0 = \Gamma_0 e^{-\Delta E/kT} \quad (2)$$

ΔE being the energy difference between 4D and 4F.

The ratio ($N_{5/2}/N_{3/2}$) is known from the experimental intensities at 5 682 Å and 5 688 Å (see Fig. 2)

$$\frac{N_{5/2}}{N_{3/2}} = \frac{5}{6} \frac{I_\pi(5\,688) + 2 I_\sigma(5\,688)}{I_\pi(5\,682) + 2 I_\sigma(5\,682)} - \frac{1}{6}. \quad (3)$$

(The quantity $I_\pi + 2 I_\sigma$ does not depend on the polarization of the excited state.) In order to measure the collisional transfer from 4D to 4F, an obvious method would be to measure the collision induced 4F-3D transition. This is difficult because the transition lines in the infrared range. We have preferred to detect the 3D-3P transitions at 8 183 Å and 8 195 Å (see Fig. 1). These transitions can also be observed without any collisions because they appear in the cascade 4D → 4P → 3D → 3P. The probability of such a cascade is small and the intensity of the 3D-3P transition is very sensitive to the pressure of the foreign gas.

Finally, we deduce the value of G_0 and $\frac{3}{2}\varphi^0$ from the intensities at 5 682 Å, 5 688 Å, 8 183 Å and 8 195 Å for both polarizations I_π and I_σ versus the pressure of noble gas.

G^0 and $\frac{3}{2}\varphi^0$ can be expressed using the cross-sections σ^0 and $\frac{3}{2}\Sigma^0$ defined by (2) :

$$G^0 = n\bar{v}\sigma^0 \quad (4)$$

$$\frac{3}{2}\varphi^0 = n\bar{v}\frac{3}{2}\Sigma^0 \quad (5)$$

n is the number of perturbers per unit volume and $\bar{v}$ is the mean relative velocity between sodium and noble gas atoms. The experimental values of σ^0 and $\frac{3}{2}\Sigma^0$ are reported on tables I and II.

For $\frac{3}{2}\Sigma^0$, the value obtained in the neon case is smaller than our previous determination [4], where we did not take into account the problem of polarization and where we used $I_\pi + I_\sigma$ instead of $I_\pi + 2 I_\sigma$ in (3).

$$\frac{d}{dt} \begin{bmatrix} \frac{5}{2}\rho_q^2 \\ \frac{3}{2}\rho_q^2 \end{bmatrix} = \begin{bmatrix} -\frac{1}{\tau} - \frac{5}{2}\Phi^2 & \frac{3}{2}\Phi^2 \\ \frac{3}{2}\Phi^2 & -\frac{1}{\tau} - \frac{3}{2}\Phi^2 \end{bmatrix} \begin{bmatrix} \frac{5}{2}\rho_q^2 \\ \frac{3}{2}\rho_q^2 \end{bmatrix} + \begin{bmatrix} \frac{5}{2}\lambda_q^2 \\ 0 \end{bmatrix} \quad (6)$$

(2) $\frac{3}{2}\Sigma^0$ is related to the usual cross-sections $\sigma(5/2 \rightarrow 3/2)$ and $\sigma(3/2 \rightarrow 5/2)$ by the relations :

$$\sigma(5/2 \rightarrow 3/2) = \sqrt{\frac{2}{3}} \frac{3}{2}\Sigma^0$$

$$\sigma(3/2 \rightarrow 5/2) = \sqrt{\frac{3}{2}} \frac{3}{2}\Sigma^0$$

(of course $\frac{3}{2}\Sigma^0$ has the same symmetry as $\frac{3}{2}\varphi^0$: $\frac{3}{2}\Sigma^0 = \frac{5}{2}\Sigma^0$).

TABLE I

Experimental value of the σ^0 cross-sections (4D-4F transfer)

	He	Ne	Ar	Kr	Xe
σ^0	4.7 ± 1.3	2.2 ± 0.5	1.3 ± 0.4	1.9 ± 0.4	1.8 ± 0.5

The values are given in 10^2 atomic units. The experimental temperature is 270 °C.

TABLE II

Experimental values of the $\frac{3}{2}\Sigma^0$ cross-sections (transfer of population)

	He	Ne	Ar	Kr	Xe
$\frac{3}{2}\Sigma^0$	6.3 ± 1.5	7.0 ± 1.3	9.6 ± 1.3	14.5 ± 1.3	22.9 ± 4.2

The usual cross-sections $\sigma(5/2 \rightarrow 3/2)$ and $\sigma(3/2 \rightarrow 5/2)$ can be deduced from $\frac{3}{2}\Sigma^0$:

$$\sigma(5/2 \rightarrow 3/2) = \sqrt{\frac{2}{3}} \frac{3}{2}\Sigma^0 \quad \text{and} \quad \sigma(3/2 \rightarrow 5/2) = \sqrt{\frac{3}{2}} \frac{3}{2}\Sigma^0.$$

The values are given in 10^2 atomic units. The experimental temperature is 270 °C.

3. Relaxation of the polarization. — In absence of collisions, the fluorescent light emitted from the 4D_{3/2} and 4D_{5/2} levels after a two-photon excitation is polarized. (The polarization rate and the principal axes of polarization depend on the polarization of the exciting light.) For instance, without a noble gas, the 5 688 Å of figure 2 is highly polarized : the polarization ratio T which is equal to

$$T = \frac{I_\sigma - I_\pi}{I_\sigma + I_\pi}$$

has the value 0.75 [10]. In presence of noble gases, the polarization rate decreases. The destruction of the light polarization is related to the relaxation rate of the alignment (which is the component $J\rho_q^2$ ($-2 \leq q \leq 2$) of rank 2 of the density matrix of the excited state [11]). The equations for the alignment are similar to equations (1) obtained for the populations (3). They are :

$\frac{5}{2}\lambda_q^2$ is the pumping rate when the frequency of the laser corresponds to the 3S_{1/2}-4D_{5/2} transition. $\frac{3}{2}\Phi^2$ describes the transfer of alignment between the 4D_{5/2} and 4D_{3/2} levels.

(3) We have not taken into account the 4F because the transfer of alignment is smaller than the transfer of population and the transfer of population between 4D and 4F is already rather small.

If $\frac{3}{2}\Phi^2$ is different from 0, it means that the collision-induced line at 5 682 Å of figure 2 is polarized. We have tried to observe such an effect. The measurements have to be performed at a low noble gas pressure because if the pressure is too high, the collisions completely depolarize both lines at 5 682 and 5 688 Å.

FIG. 3. — Polarization rates on the fluorescent line at 5 688 Å and on the collision induced line at 5 682 Å when the frequency of the laser is locked on the 3S_{1/2}-4D_{5/2} two-photon transition in sodium. The experiment is performed with the same amount of foreign gas : in the case of argon the collision induced line is not polarized. whereas for neon there is an important polarization.

Figure 3 shows the experimental results obtained in the case of argon and neon. In the case of neon, the collision-induced line is clearly polarized even if the polarization ratio of the 5 682 line is about six times smaller than the polarization ratio of the 5 688 line. In the case of argon, the polarization rate of the collision-induced line is too small to be measured with our experimental set-up.

In fact we have observed that the collision induced line is polarized in the case of helium and neon perturburbers. For the case of the heavier noble gases, no polarization has been observed.

We have carried out systematic measurements of the polarization of the fluorescent light *versus* the noble gas pressure. From these measurements, we have deduced the variation of the $\frac{J}{J}\Phi^2$ quantities with pressure. Using equation (5), we can define three cross sections $\frac{3}{2}\Sigma^2$, $\frac{5}{2}\Sigma^2$ and $\frac{3}{2}\Sigma^2$. Their experimental values are reported on table III.

4. Comparison with theoretical predictions. — It is possible, if one knows the interaction potential between the sodium atom and the noble gas, to calculate the various cross-sections presented above. Adiabatic

TABLE III
Experimental values of the $\frac{J}{J}\Sigma^2$ cross-sections
(relaxation of alignment, transfer of alignment)

	He	Ne	A	Kr	Xe
$\frac{3}{2}\Sigma^2$	17.0 ± 2.3	15.2 ± 1.5	26.2 ± 3.4	38.7 ± 4.8	51.5 ± 3.9
$\frac{5}{2}\Sigma^2$	14.7 ± 2.1	14.6 ± 1.5	23.5 ± 3.1	32.8 ± 3.7	47.8 ± 6.4
$\frac{3}{2}\Sigma^2$	1.1 ± 0.2	1.1 ± 0.2	0 ± 0.4	0 ± 0.4	0 ± 0.5

The values are given in 10² atomic units. The experimental temperature is 270 °C.

potentials for these systems have been recently calculated by Pascale and Vandeplanque [12]. We have made calculations using these potentials, but because the wavefunctions have not been published, we have used atomic wavefunctions. The atomic wavefunctions are certainly wrong at small internuclear distances, nevertheless the cross-sections are rather large. Furthermore, Pascale [13] has calculated the $\sigma(5/2 \rightarrow 3/2)$ cross-sections using the exact wavefunctions ; his results are practically identical to ours. The main problem with the atomic wavefunctions is that they do not allow the calculation of the 4D-4F transfer cross-section to be performed.

Due to the small magnitude of the spin-orbit coupling in the 4D level, (compared to 1/τ, τ being the characteristic time of a collision) we have made the computation for an atom without spin and we have recoupled the spin after the collision. We have used the straight path trajectory model with the mean velocity

$$\bar{v} = \sqrt{\frac{8 k T}{\pi \mu}},$$

(μ being the reduced mass).

The calculated values of the cross-sections are

TABLE IV
Theoretical values of the cross-sections

	He	Ne	A	Kr	Xe
$\frac{3}{2}\Sigma^0$	9.8	16.7	15.2	17.6	20.6
$\frac{3}{2}\Sigma^2$	21.2	38.1	30.0	37.3	43.0
$\frac{5}{2}\Sigma^2$	18.8	33.5	30.5	36.1	42.5
$\frac{3}{2}\Sigma^2$	+ 1.8	+ 2.3	+ 0.1	+ 0.4	− 0.1

These values are obtained using the adiabatic potentials of Pascale and Vandeplanque and atomic wavefunctions. The computation is performed for the mean relative velocity $v = \sqrt{8 k T / \pi \mu}$, T being the experimental temperature. The values are given in 10² atomic units.

reported on table IV. They can be compared with the experimental values of tables II and III. There is a large disagreement between the experimental and theoretical values in the case of neon. In this case,

the potentials [12] are certainly not precise enough. The agreement is much better for the other nobles gases. The difference between the experimental and theoretical values for ${}_{5/2}^{3/2}\Sigma^0$ in the helium case, can certainly be explained by the coupling with the 4F level which has not been taken into account in our calculation. It will be noticed that the theory predicts the transfer of alignment with the right order of magnitude and the right sign.

In the same calculation, we have also obtained the values of the shift and broadening of the 3S-4D two-photon line. The comparison with the experimental results [3] will be discussed in a more detailed paper.

5. Conclusion. — We have summarized the principal features of recent experiments [10] dealing with the application of Doppler-free two-photon excitation to the investigation of collisional problems. We have been able to measure various cross-sections, and we have compared them with theoretical predictions. More details concerning the experiment and the calculation will be presented in a more complete paper.

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References

- [1] For a recent review on Doppler-free two-photon spectroscopy see
GRYNBERG, G. and CAGNAC, B., *Rep. Prog. Phys.* **40** (1977) 791.
- [2] BIRABEN, F., CAGNAC, B. and GRYNBERG, G., *J. Physique* **36** (1975) L 41.
- [3] BIRABEN, F., CAGNAC, B., GIACOBINO, E. and GRYNBERG, G., *J. Phys. B* **10** (1977) 2369.
- [4] BIRABEN, F., CAGNAC, B. and GRYNBERG, G., *C.R. Acad. Sci.* **280B** (1975) 235.
- [5] HANSCH, T. W., HARVEY, K., MEISEL, G. and SHAWLOW, A. L., *Opt. Commun.* **11** (1974) 50.
- [6] BIRABEN, F., CAGNAC, B. and GRYNBERG, G., *C.R. Acad. Sci.* **279B** (1974) 51 and *Phys. Lett.* **48A** (1974) 469.
- [7] BIRABEN, F., CAGNAC, B. and GRYNBERG, G., *Phys. Rev. Lett.* **32** (1974) 643 and *Phys. Lett.* **49A** (1974) 71.
- [8] LEVENSON, M. D. and BLØMBERGEN, N., *Phys. Rev. Lett.* **32** (1974) 645.
- [9] CAGNAC, B., GRYNBERG, G. and BIRABEN, F., *J. Physique* **34** (1973) 845.
- [10] BIRABEN, F., Thèse Paris (1977).
- [11] OMONT, A., *Prog. Quantum Electron.* **5** (1977) 69.
- [12] PASCALE, J. and VANDEPLANQUE, J., *J. Chem. Phys.* **60** (1974) 2278.
- [13] PASCALE, J., To be published.