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INVESTIGATION OF THE LINE $\lambda = 587 \text{ nm}$ ($1s_4-2p_2$) OF KrI USING STIMULATED EMISSION IN AN ATOMIC BEAM

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Résumé. — Une méthode qui permet d'étudier dans un jet atomique de gaz rare les raies ayant un niveau inférieur résonnant est présentée : une inversion de population est produite à l'aide d'un premier laser entre un niveau excité et le niveau résonnant et un second laser accordable induit des transitions entre ces niveaux. L'expérience a été réalisée sur un jet de krypton ; l'amplitude et la largeur du signal de résonance sont calculées et sont comparées aux résultats expérimentaux. La méthode est appliquée à la mesure de déplacements isotopiques et donne :

$$\nu(^{86}\text{Kr}) - \nu(^{84}\text{Kr}) = (118 \pm 6) \text{ MHz} \quad \text{et} \quad \nu(^{84}\text{Kr}) - \nu(^{82}\text{Kr}) = (108 \pm 6) \text{ MHz}$$

pour la raie $\lambda = 587 \text{ nm}$ ($1s_4-2p_2$) de KrI.

Abstract. — A method which allows the investigation, in an atomic beam, of noble-gas lines having a resonant lower level is discussed : a population inversion is produced by means of a first laser between an excited level and the resonant level of interest ; a second tunable laser induces transitions between those two levels. The experiment has been done on a Kr beam ; the expected amplitude and width of the resonance have been calculated and are compared with the experimental data. The method has been applied to isotope-shift measurements and gives :

$$\nu(^{86}\text{Kr}) - \nu(^{84}\text{Kr}) = (118 \pm 6) \text{ MHz} \quad \text{and} \quad \nu(^{84}\text{Kr}) - \nu(^{82}\text{Kr}) = (108 \pm 6) \text{ MHz}$$

in the line $\lambda = 587 \text{ nm}$ ($1s_4-2p_2$) of KrI.

1. Introduction. — In a previous paper [1], we have described a method allowing the study of noble-gas transitions between metastable levels (i.e. levels with $J = 0$ and $J = 2$ of the $np^5(n+1)s$ configuration) and more excited levels. An atomic beam containing a certain number of atoms in metastable levels was produced by means of an electrical (hollow cathode or radiofrequency) discharge burnt inside the oven of the apparatus ; an optical-absorption experiment using a CW single-frequency dye laser was performed from the metastable levels and the fluorescence light from the laser-excited atoms was detected. The experiment was restricted to those lines having a metastable lower level, because the resonant levels are not normally populated in the atomic beam.

In this paper we report on a new method which allows the spectroscopic investigation of noble-gas transitions having a resonant lower level (i.e. one of the $J = 1$ levels of the $np^5(n+1)s$ configuration). The principle of our new experiment is given in section 2 ; a quantitative evaluation of the width and amplitude of the expected resonance signal is presented in section 3 and the results are compared

to the experimental data ; finally, in section 4 we show how the method can be applied to isotope-shift measurements and we report preliminary values of the isotope shifts of ^{82}Kr , ^{84}Kr and ^{86}Kr in the line $\lambda = 587 \text{ nm}$ of KrI.

2. Principle of the experiment. — The principle of the experiment relies on the possibility of creating a population inversion between a resonant level (level with $J = 1$) of the $np^5(n+1)s$ configuration and a level of a more excited configuration. This can be achieved as follows (Fig. 1a) :

i) By means of a first laser beam (laser 1), a level with $J = 1$ or 2 of the $np^5n'p$ configuration (level e) can be populated from a metastable level (level m) (cf. Ref. [1]).

ii) The level e decays by spontaneous emission towards several levels of the $np^5(n+1)s$ configuration. The two resonant ($J = 1$) levels are thus populated but, due to their very short lifetimes, their population is much smaller than that of the excited level. One can take advantage of this population inversion to investi-

FIG. 1. — a) Characteristics of the energy levels and of the laser fields involved in the experiment; b) Characteristics of the two-level model used in the calculation.

gate the line $r - e$ by means of a second laser beam (laser 2) which is brought into resonance with this line (Fig. 1a). The induced emission produced by this irradiation reduces the population of level e ; the population depletion can be monitored by detecting the fluorescence light emitted from level e . By scanning the laser-2 frequency, a *negative* peak centred at the optical frequency Ω_{er} (neglecting light shifts) should be recorded. In contrast to the previous laser absorption experiment of [1], this method can be considered as a laser stimulated-emission experiment.

3. Characteristics of the resonances. — **3.1 THEORETICAL EXPRESSION.** — Since we detect the fluorescence light from the atoms in level e , the quantity of interest is the population of the upper level N_e . The system under study can be considered as an ensemble of three-level atoms submitted to two monochromatic radiations quasis resonant respectively with the transition $m-e$ and with the transition $r-e$. These atoms are located inside the volume τ , the intersection of the atomic beam and of the two laser beams. If the intensities and the frequencies of the two lasers are fixed, a steady state exists for which the populations of the levels m , r and e inside volume τ remain constant.

If one takes into account the limited interaction time between the atoms and the two laser beams, it is rather difficult to calculate the steady-state values of the populations and coherences of the three-level system. Since in this work we need an evaluation of the intensity and of the width of the peak rather than a precise determination of the resonance

shape, we will replace the actual problem by a much simpler one. Namely, we will deal with a two-level system (levels e and r) submitted to a monochromatic radiation, the level e being excited at a constant rate λ by pumping from the metastable state m (Fig. 1b). Furthermore the levels e and r will be considered as non-degenerate.

The atoms are submitted to the electric field of the monochromatic wave of frequency Ω due to laser 2 ⁽¹⁾:

$$\mathbf{E}(t) = \varepsilon E_0 \cos \Omega t.$$

We define the quantity V , proportional to the electric-dipole matrix element between states e and r , by:

$$V = \frac{-eE_0}{2\hbar} \langle e | \varepsilon \cdot r | r \rangle; \quad V \text{ is supposed to be real.}$$

For an electric-dipole transition and using the rotating-wave approximation the optical Bloch equations are:

$$\begin{aligned} \dot{N}_e &= -\Gamma_e N_e - 2bV + \lambda \\ \dot{N}_r &= -\Gamma_r N_r + \gamma_{er} N_e + 2bV \\ \dot{a} &= -\frac{\Gamma_e + \Gamma_r}{2} a - \Delta b \\ \dot{b} &= \Delta a - \frac{\Gamma_e + \Gamma_r}{2} b + V(N_e - N_r). \end{aligned} \quad (1)$$

In these equations N_e , N_r , a and b are real quantities related to the matrix elements of the density operator in the following way:

$$N_e = \rho_{ee}; \quad N_r = \rho_{rr}; \quad (a + ib) = \rho_{re} e^{i\Omega t}$$

Γ_e and Γ_r are the radiative relaxation rates of levels e and r respectively, γ_{er} is the spontaneous emission rate for the $e - r$ transition and $\Delta = \Omega - \Omega_{er}$ is the detuning of the laser from the atomic-transition frequency Ω_{er} .

Solving equations (1) for the steady-state regime we find:

$$N_e = \lambda \left[\Gamma_e + \frac{4V^2}{\Gamma_e + \Gamma_r} \times \frac{(1 - \gamma_{er}/\Gamma_r)}{1 + 4\Delta^2/(\Gamma_e + \Gamma_r)^2 + 4V^2/\Gamma_r(\Gamma_e + \Gamma_r)} \right]^{-1}. \quad (2)$$

3.2 APPLICATION TO THE CASE OF Krl : CALCULATED AND EXPERIMENTAL VALUES OF THE INTENSITY AND WIDTH OF THE SIGNAL. — The experiment has been carried out on the transition $\lambda = 587 \text{ nm}$ ($2p_2 - 1s_4$) of Krl. The atoms are excited from the metastable $1s_5$ level (level m) to the level $2p_2$ (level e) and can decay towards the levels $1s_5$, $1s_4$ (level e) and $1s_2$.

⁽¹⁾ In this calculation, we use notation similar to that of Feneuille, Schweighofer and Oliver [3].

The numerical values of the relaxation and spontaneous emission have been computed by M. Aymer [2] to be :

$$\Gamma_e = 38 \times 10^6 \text{ s}^{-1}; \quad \Gamma_r = 260 \times 10^6 \text{ s}^{-1};$$

$$\gamma_{er} = 1.4 \times 10^6 \text{ s}^{-1}.$$

The laser power per unit area being 30 mW/mm^2 , one finds $V = 35 \times 10^6 \text{ s}^{-1}$. According to eq. (2), the relative decrease of the fluorescence signal at resonance is : $(N_e^0 - N_e^M)/N_e^0 = 27\%$; N_e^0 (or N_e^M) is the population of level e when laser 2 is off (or on).

The width at half maximum (i.e. for $N_e = \frac{N_e^0 + N_e^M}{2}$) of the peak is 57 MHz.

Figure 2 shows a recording of the fluorescence signal obtained by varying Ω around the optical frequency Ω_{er} ($\lambda = 587$ nm). The laser 1 was resonant with the transition $1s_5-2p_2$ ($\lambda = 556$ nm) of ^{84}Kr , so that only ^{84}Kr atoms are excited to the level e . The atomic beam apparatus and the first laser ($\lambda = 556$ nm) in use were the same as in our previous experiment [1]; the second laser beam was delivered by a jet stream dye laser (Spectra Physics Model 580) and was nearly collinear to the first laser beam, i.e. perpendicular to the atomic beam. To avoid difficulties arising from slow intensity and frequency fluctuations of laser 1, the intensity of laser 2 was mechanically modulated (200 Hz) and the fluorescence signal was detected

using lock-in amplification techniques. On figure 2 the relative decrease of the signal at resonance is about 7%; this number has to be multiplied by a factor of 2 because the laser 2 is only efficient half of the time due to the modulation. The width of the peak is about 70 MHz. The agreement between these experimental results and the calculations is quite satisfactory considering the approximations.

4. Measurement of isotope shifts. — As described above the method cannot be applied to isotope-shift measurements because only one isotope is excited by laser 1 to level e at a time. If laser 1 is made to cross the atomic beam not at a right angle but at an angle θ and is reflected back on itself (Fig. 3) an atom with velocity v is in fact submitted to two radiations with wavenumbers

$$\sigma = \sigma_1(1 + v \cos \theta/c) \text{ and } \sigma' = \sigma_1(1 - v \cos \theta/c);$$

FIG. 3. — Application of the method to isotope-shift measurements. The directions of the two laser beams and of the atomic beam are in the same plane. The fluorescence emitted in the direction perpendicular to this plane is passed through a colour filter which isolates the line $2p_2-1s_2$ ($\lambda = 826$ nm) (cf. Fig. 1a), thus eliminating the stray light from the lasers.

FIG. 2. — Experimental resonance curve : a) Only ^{84}Kr is excited by laser 1. Trace α shows the evolution of the fluorescence emitted from the excited level with the laser 2 frequency and trace β shows only the part of this fluorescence which is modified by laser 2 (amplitude modulation of laser 2 and lock-in amplification on use); b) Both ^{84}Kr and ^{86}Kr are excited by laser 1 to allow the isotope-shift measurement. Calibration fringes are provided by a temperature-controlled spherical Fabry-Pérot interferometer.

σ_1 being the laser 1 wavenumber. By adjusting σ_1 and θ , laser 1 can be Doppler-tuned into resonance with the transition $m-e$ of two isotopic species for atoms in the beam having the average velocity v_m .

The adjustment of the angle θ , whose value depends both upon the isotope shift of the transition $m-e$ and upon v_m , has been made by recording the fluorescence intensity when the laser 1 frequency was tuned the laser 2 being off. For the isotope pair $^{86}\text{Kr}-^{84}\text{Kr}$ the isotope shift of the transition $m-e$ ($\lambda = 556$ nm) is 86 MHz [1]; we have found $\theta = 78.5^\circ$, this value corresponds to a temperature of about 160 K for

the metastable atoms inside the *oven*. This temperature seems reasonable in view of the fact that the discharge tube (the *oven*) is cooled with liquid nitrogen.

By scanning laser 2, which crosses the atomic beam at right angles (Fig. 3), it is then possible to measure the isotope shift of pairs of isotopes in the line *r-e*. Figure 2*b* shows a typical recording obtained with the isotope pair ^{86}Kr - ^{84}Kr . For the shifts ^{86}Kr - ^{84}Kr and ^{84}Kr - ^{82}Kr of the $\lambda = 587 \text{ nm}$ ($1s_4$ - $2p_2$) KrI line we have obtained

$$\delta\nu(86 - 84) = (118 \pm 6)\text{MHz}$$

and

$$\delta\nu(84 - 82) = (108 \pm 6)\text{MHz}^{(2)}.$$

Combining these shifts with those recently measured [1] for the line $\lambda = 556 \text{ nm}$, we obtain the difference of the isotope shifts of the two levels $\nu(1s_5)$ - $\nu(1s_4)$, namely $(32 \pm 8)\text{MHz}$ for the pair ^{86}Kr - ^{84}Kr and $(25 \pm 8)\text{MHz}$ for the pair ^{84}Kr - ^{82}Kr . These values are in excellent agreement with those obtained by Jackson ⁽³⁾ from the study of infrared lines of KrI, respectively 31 and 28 MHz, and are

⁽²⁾ The error margins are equal to the mean square error of about 10 recordings. These figures are not very reliable since systematic errors may arise from incomplete resolution [cf. Fig. 2*b*].

⁽³⁾ D. A. Jackson, Private communication.

probably due mainly to a difference between the specific-mass shifts of the two levels $1s_5$ and $1s_4$ arising from departure from the LS coupling scheme [4].

5. Conclusion. — We have shown that it is possible with an atomic beam to turn from the usual laser-absorption technique using fluorescence for detection to a laser stimulated-emission experiment in order to study hyperfine and isotopic structures of transitions involving, as lower levels, resonance levels which have very short lifetimes and are normally not populated. This is particularly interesting for the krypton line we have investigated, because other methods of Doppler-free spectroscopy do not seem to have succeeded in this case (see e.g. [5]).

Several technical improvements would be necessary to increase the signal-to-noise ratio in our experiment, among which a better long-term stabilization of the amplitudes and frequencies of the lasers is the most important. The use of enriched samples would also be necessary to investigate the rarer isotopes of krypton.

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