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Optical pumping and g-factor of the $^3P_0$ state of the first excited configuration of rare gases odd isotopes

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Résumé. — Les métastables np$^5(n + 1)s$ $^3P_0$ (1s$^3$ en notation de Paschen) d’isotopes impairs du néon, du krypton et du xénon ont été pompés optiquement dans un jet atomique grâce à un laser à colorant continu monomode. Une expérience de résonance magnétique conduit à la mesure du facteur de Landé de l’état $^3P_0$: $3.02(2) \times 10^{-4}$ pour $^{21}$Ne; $0.974(4) \times 10^{-4}$ pour $^{83}$Kr; $7.781(2) \times 10^{-4}$ pour $^{129}$Xe et $-2.305(1) \times 10^{-4}$ pour $^{131}$Xe. Ces résultats sont comparés à des prévisions théoriques.

Abstract. — Metastable np$^5(n + 1)s$ $^3P_0$ (1s$^3$ in Paschen notation) atoms of odd isotopes of neon, krypton and xenon have been optically pumped in a beam of metastable atoms by means of a C.W. single-mode dye laser. A magnetic resonance experiment leads to the determination of the g-factor of the $^3P_0$ state: $3.02(2) \times 10^{-4}$ for $^{21}$Ne; $0.974(4) \times 10^{-4}$ for $^{83}$Kr; $7.781(2) \times 10^{-4}$ for $^{129}$Xe and $-2.305(1) \times 10^{-4}$ for $^{131}$Xe. These results are compared with theoretical predictions.

1. Introduction.

For odd isotopes, the $J = 0$ levels of atoms may have magnetic moments significantly different from the pure nuclear values. This effect is due to hyperfine and Zeeman coupling which mixes nonzero $J$ contributions into the wave functions describing $J = 0$ states and therefore introduces some electronic magnetism into these states. Experimental evidence of such effects has already been reported in mercury [1], cadmium [2] and neon [3]. We describe here a magnetic resonance experiment on atoms in the np$^5(n + 1)s$ $^3P_0$ metastable states of $^{21}$Ne, $^{83}$Kr, $^{129}$Xe and $^{131}$Xe using a laser-optical-pumping-atomic-beam-magnetic-resonance technique, leading to the determination of the Landé factors of the $^3P_0$ states.

2. Experiment.

2.1 Generalities. — For krypton and xenon atoms, it is probably impossible to perform a magnetic resonance experiment on the $^3P_0$ state in a cell, as we have done for neon [3]: the $^3P_0$ state relaxation in a cell is too fast in the case of the heavy rare gases [4]. So, we have used an atomic beam containing metastable atoms, for which the magnetic resonance width is determined by the interaction time of the atoms with the radiofrequency field.

The experimental set up is shown in figure 1. Two laser beams interact with the atomic beam. The first one, the pumping beam, induces population differences between Zeeman sublevels of the $^3P_0$ metastable level. The second one, the detection beam, probes these differences. Between the two interaction regions, atoms are submitted to a steady magnetic field and to a radiofrequency field, which may induce transitions between $^3P_0$ Zeeman sublevels. Such a method, where the pumping region is spatially sepa-

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(*) L.A. au C.N.R.S. n° 19.
rated from the detection one, is very sensitive to any change of the Zeeman sublevel populations occurring in the light free region [5], so that it is most suitable for magnetic resonance experiments.

2.2 ATOMIC BEAM. — The atomic beam apparatus, built in the Laboratory, is derived from that of Stanley [6]. The vacuum tank is divided into two chambers which are separately pumped by two diffusion pumps. Two circular apertures, 1.5 mm in diameter, 32 mm apart, ensure the collimation of the atomic beam. The first one is at the end of the tube through which the gas is injected into the first vacuum chamber, the second one is at the separation between the two chambers. With this system, the major part of the atoms leaving the injection tube are evacuated by the first pump, so that a good vacuum (a few 10⁻⁷ torr) can be obtained in the second chamber where the optical pumping of the metastable atoms and the magnetic resonance take place.

The atoms are carried into the metastable state by means of an electrodeless U.H.F. discharge at the end of the injection tube. In the case of neon, we have used samples enriched to 10% in ¹⁷⁰Ne so that the gas had to be recirculated: from the exit of the primary pump, it was readmitted into the injection tube after passing through a liquid nitrogen cooled trap in order to remove the impurities. This procedure is not necessary with xenon and krypton, where the natural gas contains odd isotopes in sufficient abundance to be used in our experiments (26.4% ¹²⁹Xe and 21.2% ¹³¹Xe in natural xenon, 11.5% ⁸³Kr in natural krypton).

2.3 LASER. — The optical pumping of the metastable ³P₀ atoms (¹s₈ in Paschen notation) is performed with a C.W. single-mode dye laser tuned on one of the hyperfine components of an allowed ¹s₈-2P₁ transition (see energy levels diagram and pumping lines on Fig. 2). Single-mode operation is achieved by means of a double Michelson selector [7]. The branching ratios of the pumping lines being noticeably smaller than 1 (~ 40% for Ne and ~ 50% for Kr and Xe), the optical pumping proceeds chiefly by a selective depletion of the Zeeman sublevels of the ³P₀ state rather than by Kastler's original scheme. The detection beam comes from the same dye laser as the pumping beam; both are π polarized and propagate perpendicularly to the atomic beam and to the steady magnetic field H₀ directions, at a distance of 20 cm from each other. The fluorescence light, induced by the detection beam, is observed perpendicularly to the atomic beam and to the pumping beam directions with a photomultiplier PM2 (EMI 9558 QB), the anode current of which is fed into a lock-in amplifier. (The photomultiplier PM1 of Fig. 1 is used only for the control of the fluorescence light induced by the pumping beam). The laser beam powers are of the order of 10 mW for the pumping beam, 0.5 mW for the detection beam, and their cross sections are about 20 mm².

2.4 MAGNETIC RESONANCE. — The steady magnetic field H₀ is produced by a pair of Helmholtz coils which are fed by a stabilized current generator 60 V-20 A, giving a maximum field intensity of 115 G. The radiofrequency field Hᵣ is produced by a cylindrical coil 12 cm in length, 5 cm in diameter, which belongs to a resonant circuit. Resonant frequency is in the 12-115 kHz range, depending on the tuning capacity. Hᵣ rotating field intensities used for magnetic resonance are smaller than 3 gauss for ¹⁷⁰Ne, 2 gauss for ⁸³Kr, 1 gauss for ¹²⁹Xe and 1.2 gauss for ¹³¹Xe. We have verified in each case that the corresponding Bloch Siegert shift is at least ten times smaller than the statistical error of our measurements. The radiofrequency field is chopped at 35 Hz: this frequency is the reference frequency of the lock-in amplifier.

Fig. 2. — First excited configurations and transitions involved in the experiments. ⇒ Laser wavelength (in Å); → Fluorescence wavelength (in Å).
the output signal of which is recorded versus the steady magnetic field $H_0$, which is swept around the resonance position. A signal averager is used to improve the signal-to-noise ratio.

3. Theory.

In our previous work [3], a second-order perturbation theory was developed, in which we kept only the terms linear with respect to the magnetic field, leading to the expression:

$$g(3P_0) = g(1S_0) + 2 \sum_{s=0,1} \langle 3P_0, I, m | H_h | 2S+1P_1, I, m \rangle + \langle 2S+1P_1, I, m | H_z | 3P_0, I, m \rangle \times \left\{ m\mu_0 H_0 [E(3P_0) - E(2S+1P_1)] \right\}^{-1}.$$  \hspace{1em} (1)

$g(1S_0)$ is the ground state $g$-factor, $\mu_0$ the Bohr magneton. $| 3P_1, I, m \rangle$ and $| 1P_1, I, m \rangle$ are the intermediate coupling wavefunctions of the $m$ component of the $F = I$ hyperfine sublevel of the $3P_1$ and $1P_1$ levels of the $np^5(n+1)s$ configuration. $H_z$ and $H_h$ are the Zeeman and hyperfine Hamiltonians respectively. Expression (1) only takes into account the mixing of $3P_1$ and $1P_1$ states of the lowest excited configuration into the $3P_0$ state wave function; but in the case of xenon the interaction between $5p^56s$ and $5p^55d$ configurations cannot be neglected. So (1) has to be generalized into:

$$g(3P_0) = g(1S_0) + 2 \sum_{s=0,1} \langle 3P_0, I, m | H_h | a, I, m \rangle + \langle a, I, m | H_z | 3P_0, I, m \rangle \times \left\{ m\mu_0 H_0 [E(3P_0) - E_a] \right\}^{-1}.$$  \hspace{1em} (2)

$| a, I, m \rangle$ is the wave function of the Zeeman hyperfine sublevel of quantum numbers $J = 1$, $F = I$, $m$ of level $a$ with energy $E_a$. It is a linear combination of the pure LS coupling wavefunctions of the $J = 1$ and $F = I$ levels of the $np^5(n+1)s$ and $np^5nd$ configurations.

The $a$ sum runs over all the $J = 1$ levels of these two configurations. In the same way $| 3P_0, I, m \rangle$ is the wave function of the Zeeman hyperfine $F = I$, $m$ sublevel of the lowest excited $J = 0$ level, which is a linear combination of the two pure LS coupling wavefunctions $| 3P_0, I, m \rangle$ belonging to the $np^5(n+1)s$ and $np^5nd$ configurations. In the cases where one can neglect the interaction between these two configurations (neon and krypton), expressions (1) and (2) are identical.

As in [3], we use here an effective Hamiltonian including relativistic effects:

$$H_h = X^{(1)} I^{(1)}$$

with

$$X^{(1)} = a_1 s_1^{(1)} + a_2 I_1^{(1)} - a_3 \sqrt{10} (s_1^{(1)} c_1^{(2)})^{(1)} + a_4 s_2^{(1)}$$

where the $a$ coefficients are the coupling constants of the $p$ hole and the $s$ electron with the nuclear spin. In operators $s_1^{(1)}, I_1^{(1)}, c_1^{(2)}$, the subscript $i = 1$ or 2 refers to the $p$ hole or to the $s$ electron respectively. $H_h$ is the magnetic dipolar part of the whole Hamiltonian, the quadrupolar part yielding zero contributions to the matrix elements involved in (1) or (2). The hyperfine coupling constants $A$ of the various levels are linear combinations of the $a$ parameters, which allows a computation of the latter by a least square fit of the observed values of the $A$'s. The $a_i$'s are given by Husson and Grandin for neon [8] and krypton [9] and by Coulombre and Sinzelle for xenon [10] (1). The expansion of the intermediate coupling wave functions used in (1) and (2) on a basis of pure LS coupling functions has been made by Liberman [11] for neon and xenon and Aymar (private communication, quoted by [9]) for krypton. This allowed us to obtain the matrix elements of $H_z$ and $H_h$ which appear in (1) and (2) from the corresponding matrix elements between pure LS coupling wavefunctions. The quoted uncertainties are twice those which arise from the inaccuracies estimated by [8-10] on the hyperfine coupling constants $a_2$, $a_3$ and $a_4$. The factor of 2 has been introduced, somewhat arbitrarily, to take into account the unknown uncertainties on $a_1$ and on the intermediate coupling wavefunctions.

The theoretical values of $g(3P_0)$ that we have thus computed are given in table I.

4. Results.

4.1 Neon. — In order to test our experimental set up, we have first performed the magnetic resonance experiment on the $2p^53s$ $3P_0$ level of $^{21}$Ne, the $g$-factor of which has already been measured [3]. The dye (rhodamine B) laser, pumped by an ionized argon laser is tuned on one of the hyperfine transitions of the $6267$ Å line: $F = 3/2 \rightarrow F' = 3/2$ or $F = 3/2 \rightarrow F' = 5/2$. We have observed the magnetic resonance at $30$ kHz. No filter is used at the detection, thus the photomultiplier receives the $4$ fluorescence transitions at $5976$, $6128$, $6267$ and $6717$ Å. Integration times of about one hour are necessary to get a signal-to-noise ratio of $20$. The magnetic resonance curves are analysed as Lorentzian shaped curves, the centre of which gives the resonant field. The mean value of the $g$-factor obtained after $72$ measurements (half with increasing, half with decreasing field) is $g(3P_0) = (3.02 \pm 0.02) \times 10^{-4}$, where the uncertainty is taken as the standard deviation of measured values.

(1) We used the parameters « nouvelle option » of reference [10], i.e. those of the last line of its table IV.
Table I. — Theoretical and experimental g-factor values of the neon, krypton and xenon odd isotopes.
(a) Result of [3]; (b) Value from the present atomic beam experiment.

The uncertainty on our experimental results is taken as the standard deviation. The sign of \( g(^3P_0) \) is from the theory, the experiment yielding only its absolute value \( |g(^3P_0)| \). For theoretical uncertainties, see end of section 3.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>( g(^{1}S_0) ) ( \times 10^4 ) and reference</th>
<th>( g_{th}(^3P_0) ) ( \times 10^4 )</th>
<th>( g_{exp}(^3P_0) ) ( \times 10^4 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{21}\text{Ne})</td>
<td>2.40146(6) [12]</td>
<td>3.024(12)</td>
<td>3.027(8)(a)</td>
</tr>
<tr>
<td>(^{83}\text{Kr})</td>
<td>1.17043(7) [13]</td>
<td>0.956(18)</td>
<td>0.974(4)(b)</td>
</tr>
<tr>
<td>(^{129}\text{Xe})</td>
<td>8.4143(3) [14]</td>
<td>7.863(160)</td>
<td>7.781(2)(b)</td>
</tr>
<tr>
<td>(^{131}\text{Xe})</td>
<td>2.49430(7) [14]</td>
<td>2.333(48)</td>
<td>2.305(1)(b)</td>
</tr>
</tbody>
</table>

This value is in very good agreement both with our previous measurement [3] and with the theoretical evaluation (see table I). Its accuracy is limited by the width of the magnetic resonance curves (half width at half height (HWHH) 8 to 10 G) due to R.F. broadening and chiefly to the finite interaction time with the R.F. field (HWHH extrapolated at zero field intensity \( \Delta H_0 = 6.7 \pm 0.8 \) G).

4.2 KRYPTON. — The dye (oxazine 725) laser pumped by an ionized krypton laser is tuned on one of the hyperfine transitions of the 7 855 Å line (\( 1s_3 \rightarrow 2p_3 \)) \( F = 9/2 \rightarrow F' = 7/2 \) or \( F = 9/2 \rightarrow F' = 11/2 \) and the fluorescence light toward \( 1s_5 \) and \( 1s_4 \) levels is observed through a Schott filter which is transparent in the 3 500-6 000 Å range, but stops the red laser light at the excitation wavelength in order to avoid additional noise due to stray light. We have observed the magnetic resonance at 12 kHz. An integration time of one hour is necessary to get a signal-to-noise ratio of about 30. The mean value of the g-factor obtained after 58 measurements is: \( g(^3P_0) = 0.974(4) \times 10^{-4} \) to be compared with the theoretical prediction of table I: \( 0.956(18) \times 10^{-4} \).

The HWHH of the resonance curves vary between 10 and 13 G depending on R.F. field intensity, with an extrapolated \( \Delta H_0 \) value of 9.0 ± 0.8 G.

4.3 XENON. — We have used the same dye laser as for krypton, tuned on the hyperfine transition \( F = 3/2 \rightarrow F' = 5/2 \) (\(^{131}\text{Xe}\)) or \( F = 1/2 \rightarrow F' = 3/2 \) (\(^{129}\text{Xe}\)) of the 7 642 Å line (\( 1s_3 \rightarrow 2p_3 \)). At the detection, the Schott filter eliminates the laser stray light. The magnetic resonance has been observed at 33 kHz and 114 kHz for \(^{131}\text{Xe}\) and \(^{129}\text{Xe}\) respectively, which corresponds to a resonant field of about 100 G. In such magnetic fields, the intervals between the Zeeman sublevels of the excited \( 2p_3 \) state are large enough for the laser to excite the atoms in only one Zeeman sublevel of one hyperfine component of the \( 2p_3 \) level. So a \( \pi \) transition may induce a population difference between the \( 1s_3 \) Zeeman sublevels even in the case of \(^{131}\text{Xe}\), the nuclear spin of which is \( I = 1/2 \). This is demonstrated in figure 3 which shows the fluorescence signal received by PM2 when one tunes the laser through the \( F = 1/2 \rightarrow F' = 3/2 \) component of the \(^{129}\text{Xe}\) 7 642 Å line in a 105 G field. Curve 3a is obtained without the pumping beam and shows the clearly resolved Zeeman structure. Curve 3b is obtained with the pumping beam on; the signal has nearly completely disappeared, which shows that, among the two \( 1s_3 \) Zeeman sublevels, the one which is pumped has been efficiently emptied. Finally, curve 3c represents the fluorescence with both pumping beam and resonant R.F. on; part of the signal has been restored, which demonstrates that, at the exit of the pumping region, there exists a noticeable population of one of the Zeeman sublevels, while the other is nearly emptied (as shown above by curve 3b).

In the case of xenon and krypton, a better detection of the laser induced fluorescence light gives a better signal-to-noise ratio than for neon, so narrower magnetic resonance curves have been obtained by lowering the R.F. field intensity. But, although the signal-to-noise ratio of the fluorescence signal is of the same order of magnitude for xenon and krypton when no static magnetic field is applied, the magnetic

![Fig. 3. — Resolved Zeeman effect of the F=1/2→F'=3/2 component of the 7 642 Å line. H_o = 105 G. \( \pi \) polarized pumping and detection beams. a) Pumping beam and radiofrequency field \( H_1 \) off; b) Pumping beam on — Radiofrequency field \( H_1 \) off; c) Pumping beam and radiofrequency field \( H_1 \) on.](image)
resonance signal is much better in the case of xenon (particularly $^{129}\text{Xe}$), for the reason that, with a static magnetic field of about 100 G, the fluorescence signal intensity in the case of krypton is reduced by the great number of Zeeman components (though these are not resolved, the interval between the extreme components is 315 MHz for $F' = \frac{7}{2}$ and 407 MHz for $F' = \frac{11}{2}$).

Moreover, the HWHH $\Delta H_0$ extrapolated to zero R.F. field intensity varies as $1/gT$ where $T$ is the transit time of the atom in the R.F. field. $T$ varies as $\sqrt{M}$, where $M$ is the atomic mass, so that $\Delta H_0$ varies as $1/(g \sqrt{M})$. That is approximately verified (2) by the observed $\Delta H_0(\text{129Xe}) = 0.8 \pm 0.1$ G, $\Delta H_0(\text{131Xe}) = 2.7 \pm 0.2$ G and by the above reported values for $^{21}\text{Ne}$ and $^{83}\text{Kr}$. The $^{21}\text{Ne}$ value is a little too high with respect to the three other ones. Sharper resonances in the case of heavy rare gases allow more accurate g-factor measurements.

For $^{131}\text{Xe}$, we have performed 38 g-factor measurements, a signal-to-noise ratio of about 30 was obtained after 15 to 30 min of integration time. For $^{129}\text{Xe}$ (54 g-factor measurements), a signal-to-noise ratio of about 40 was obtained after 10 to 20 min of integration time. The measured values of g-factors are

$$|g(\text{131Xe})| = 2.305(1) \times 10^{-4}$$
$$|g(\text{129Xe})| = 7.781(2) \times 10^{-4}$$

to be compared with the theoretical predictions of table I.

Reference [1] demonstrated, in the somewhat similar case of the $^{3}\text{P}_0$ level of $^{199}\text{Hg}$ and $^{201}\text{Hg}$, the existence of a "g-factor anomaly", with the same origin as the well known hyperfine structure anomaly: the ratio of the $^{3}\text{P}_0$ g-factors of both isotopes is not exactly the same as the corresponding ratio for the $^{1}\text{S}_0$ ground level. In the present case of the xenon isotopes, we observe:

$$\Delta \equiv \frac{129}{131} \left( \frac{g(\text{3P}_0)}{g(\text{1S}_0)} \right) - 1 = 0.0007(8)$$

which does not differ significantly from 0. Let us remark that the hyperfine structure anomaly of the metastable $^{3}\text{P}_2(1s_2)$ level of Xe is, from the atomic beam magnetic resonance results of Faust and Mac Dermott [16] and the NMR data of Brinkmann [14]:

$$\Delta' \equiv \frac{129}{131} \left( \frac{g(\text{3P}_2)}{g(\text{1S}_0)} \right) \times \frac{131}{129} \left( \frac{g(\text{1S}_0)}{g(\text{3P}_0)} \right) - 1 = 0.00055(4).$$

The "g-factor anomaly" $\Delta$ is expected to be significantly smaller than $\Delta'$ because the hyperfine coupling contribution to $g(\text{3P}_0)$ is relatively small in xenon: $(g(\text{3P}_0) - g(\text{1S}_0))/g(\text{3P}_0) \approx -8\%$. It is therefore not surprising that we could not observe a significant "g-factor anomaly".

5. Conclusion.

We have reported here the first measurement of the g-factor of the $^{3}\text{P}_0$ state in the lowest odd configuration of $^{83}\text{Kr}$, $^{129}\text{Xe}$ and $^{131}\text{Xe}$. These parameters are compared to theoretical evaluations and the agreement is good. $g(\text{3P}_0)$ differs significantly from $g(\text{1S}_0)$ because of hyperfine coupling. One theoretically predicts and, indeed, observes g-shifts of different signs in the heavy rare gases and in neon. For $^{21}\text{Ne}$, $|g(\text{3P}_0)| > |g(\text{1S}_0)|$, as already observed with mercury [1] and cadmium [2]. On the other hand, for $^{83}\text{Kr}$, $^{129}\text{Xe}$ and $^{131}\text{Xe}$ $|g(\text{3P}_0)| < |g(\text{1S}_0)|$. This simply results from a change from nearly LS coupling in neon to nearly jl coupling in the heavy gases: it turns out that the two terms in the summation of equation (1) are of opposite signs and therefore partly cancel. From figure 2, it appears that the nearest, the chief, perturber of $1s_3$ is $1s_4$ in neon but $1s_2$ in krypton and xenon, hence the change of sign of the g-factor correction.

The fact that the g-shift appears as the sum of two contributions of opposite signs is unfavourable for the calculation accuracy, which, except in the case of neon, is considerably lower than the experimental precision. This should stimulate new theoretical studies of the hyperfine coupling in the first excited configuration of rare gases, especially in the case of $^{129}\text{Xe}$ for which our experimental data are reasonably accurate: a few $10^{-4}$ for $g(\text{3P}_0)$ which corresponds to $3 \times 10^{-3}$ for the g-shift $g(\text{3P}_0)-g(\text{1S}_0)$, the significant parameter since it is proportional to the hyperfine coupling.

The possibility of using $g(\text{3P}_0)$ as a valuable test of hyperfine coupling theories as well as the prospect of measuring "g-factor anomalies" is incentive to search for a still higher experimental accuracy. This seems to be straightforward, it is just a matter of paying the price for better relative sharpness of resonance curves, i.e. higher magnetic fields $H_0$ and a longer atomic beam, in order to increase the atoms-R.F. interaction time.

\textsuperscript{(2)} Although the proportionality of observed $\Delta H_0$ to $1/(g \sqrt{M})$ is fairly good, the coefficient is significantly (1.8 times) bigger than that computed from the geometry of our apparatus, with the help of chapter V and appendix D of reference [15]. We do not know the reason of this discrepancy.
References