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Measurement of the M_1 amplitude and hyperfine mixing between the $6S_{1/2}$ - $7S_{1/2}$ caesium states

M. A. Bouchiat, J. Guéna and L. Pottier

Laboratoire de Spectroscopie Hertzienne de l'E.N.S. (*),
24, rue Lhomond, 75231 Paris Cedex 05, France

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Résumé. — Nous avons mesuré le rapport de l'amplitude M_1 à la polarisabilité vectorielle β de la transition $6S_{1/2}$ - $7S_{1/2}$ du Cs, par une méthode utilisant des effets d'interférence induits par effet Stark, exempte de difficultés liées à un fond continu. L'amplitude observée varie d'une composante hyperfine à l'autre selon $M_1 + (F' - F) M_1^{\text{hf}}$ avec

$$M_1 = -(4,13 \pm 0,18) \times 10^{-5} | \mu_B | / c$$

et

$$M_1^{\text{hf}} = -(0,763 \pm 0,066) \times 10^{-5} | \mu_B | / c.$$

M_1^{hf} résulte d'un mélange entre $6S_{1/2}$ et $7S_{1/2}$ induit par l'interaction hyperfine H^{hf} et fournit une valeur de $\langle 7S_{1/2} | H^{\text{hf}} | 6S_{1/2} \rangle$ en accord avec une prédiction théorique où intervient le produit des écarts hyperfins de 6S et 7S. La signification théorique de ce résultat est discutée.

Abstract. — We have measured the ratio M_1/β of the M_1 amplitude to the vector polarizability β of the $6S_{1/2}$ - $7S_{1/2}$ transition in Cs, by a method using Stark-induced interference effects, free from background difficulties. The amplitude varies from one hyperfine component to the other, according to $M_1 + (F' - F) M_1^{\text{hf}}$ with

$$M_1 = -(4.13 \pm 0.18) \times 10^{-5} | \mu_B | / c$$

and

$$M_1^{\text{hf}} = -(0.763 \pm 0.066) \times 10^{-5} | \mu_B | / c.$$

M_1^{hf} results from a mixing between $6S_{1/2}$ and $7S_{1/2}$ induced by the hyperfine interaction H^{hf} and yields a value of $\langle 7S_{1/2} | H^{\text{hf}} | 6S_{1/2} \rangle$ which agrees with a theoretical prediction involving the product of the 6S and 7S hfs splittings. The theoretical significance of this result is discussed.

The highly forbidden $6S_{1/2}$ - $7S_{1/2}$ transition of caesium has been under active study since it was recognized to be a good place to search for parity violation in atoms [1]. Here we report the measurement of its magnetic dipole amplitude, which turns out to be different for different hfs components, in quantitative agreement with the theoretical prediction [2]. The primary interest of this result is that it yields the off-diagonal matrix element of the hyperfine interaction

(*) Associé au C.N.R.S.

between two different $nS_{1/2}$ states. Comparison of this quantity with the accurately known hyperfine structures of the two S states tests the importance of many-body effects between electrons in the hyperfine interaction.

Hyperfine mixing between states of different principal quantum number has often been discussed for two-electron atoms and is under active study in Rydberg states of alkaline-earth elements [3]. On the contrary, concerning one-electron atomic states, we do not know any other manifestation than the present one (and the very similar case associated with the M_1 amplitude of the $6P_{1/2}$ - $7P_{1/2}$ transition of thallium [4]).

Between two $S_{1/2}$ states, the only multipole moment permitted by symmetry is the magnetic dipole M_1 . But since different S states are orthogonal, the M_1 amplitude vanishes in first approximation. Yet, weak relativistic effects such as the combined effect of core excitation and spin-orbit coupling lead to a non-zero magnetic dipole amplitude M_1 equal to a fraction of $10^{-4} |\mu_B|/c$ [2, 5]. Furthermore, the hyperfine interaction mixes S states of different principal quantum number and gives rise to a second magnetic dipole amplitude $M_1^{\text{hf}}(F' - F)$, proportional to the difference $F' - F$ of the hyperfine quantum numbers of the two sublevels involved in the transition [2]. The two contributions can be distinguished by measuring the total magnetic dipole amplitude :

$$M'_1 = M_1 + M_1^{\text{hf}}(F' - F) \quad (1)$$

in the various hfs components ($6S_{1/2}, F - 7S_{1/2}, F'$) of the line. M_1^{hf} offers the particular interest of being accurately predicted as long as many-body effects between electrons are neglected. Comparison between theory and experiment thus constitutes a test of this approximation, as will be pointed out in the conclusion of this paper.

The first observed manifestation of M_1 was an interference effect which takes place in the excitation process between M_1 and the E_1 amplitude induced by a Stark field, giving rise to an electronic polarization in the 7S state. Measuring this polarization, we obtained in 1976 the first determination of M_1 , in magnitude and sign [6]. Later on, another group measured the intensities of the hfs components with and without a Stark field. Beside a confirmation of the order of magnitude of M_1 , they obtained the first experimental determination of M_1^{hf} [7]. Surprisingly, it turned out to disagree with the theoretical prediction, supposed to be reliable. This was one reason to have a fresh look at this problem. Another reason was the availability of our apparatus, successfully used to measure a (parity violating) dipole amplitude 10^4 times smaller than M_1 [8]. Still one more reason was the existence of reliable theoretical [9] and semi-empirical [10] values recently obtained by various groups for the scalar polarizability α of the 6S-7S transition as well as accurate measurements [11] of the ratio α/β . The vector polarizability β is necessary to extract M_1 and M_1^{hf} from the directly measured quantities M_1/β and M_1^{hf}/M_1 .

The 6S-7S transition amplitude can be computed from the effective dipole operator [2]

$$\mathbf{d} = M'_1 \boldsymbol{\sigma} \times \hat{k} - \alpha \mathbf{E} - i\beta \boldsymbol{\sigma} \times \mathbf{E} \quad (2)$$

where $\boldsymbol{\sigma}$ is the Pauli operator of the electronic spin and $\hat{k}$ the propagation direction of the laser beam. The second and third terms are the scalar and vector components of the electric dipole induced by the Stark field $\mathbf{E}$. The experiment basically consists in measuring an electronic polarization $\mathbf{P}^{(1)}$ arising in the 7S state from interference between the first and the second or third terms of (2) when one hfs component of the transition is excited with a laser beam polarized linearly along $\mathbf{E}$ ($\alpha E \cdot M'_1$ interference) or $\perp \mathbf{E}$ ($\beta E \cdot M'_1$ interference). This polarization $\mathbf{P}^{(1)}$, directed along $\hat{k} \times \mathbf{E}$, is odd under reversal of either $\hat{k}$ or $\mathbf{E}$. The configuration is shown in figure 1a. The magnitude of $P^{(1)}$, in the different hfs components is given in table I. To eliminate possible depolarizations $^{(1)}$ (from collisions or in the optics), we calibrate with a large electronic

⁽¹⁾ The polarization loss amounts typically to 10 to 15 %, as deduced from the calibration procedure presented here.

Fig. 1. — a) Experimental configuration. $\mathbf{E}$: Stark field; $\hat{k}$: laser direction; $\mathbf{P}^{(1)}$ and $\mathbf{P}^{(2)}$: components of the $7S_{1/2}$ state electronic polarization. b) Set-up. P : polarization modulator; M : tilted mirror; $\lambda/4$: rotating quarter-wave plate; A : polaroid analyser; D : detector; LI : lock-in amplifiers.

Table I. — Electronic polarization $P^{(1)}$ created by $\alpha E \cdot M'_1$ or $\beta E \cdot M'_1$ interference in the excited state of the $(6S, F \rightarrow 7S, F')$ hfs component. ϵ is the incident plane polarization.

	$4 \rightarrow 4$	$3 \rightarrow 3$	$3 \rightarrow 4$	$4 \rightarrow 3$
ϵ	$\parallel \mathbf{E}$	$\parallel \mathbf{E}$	$\perp \mathbf{E}$	$\perp \mathbf{E}$
$P^{(1)}$	$\frac{5}{6} \frac{M_1}{\alpha E}$	$\frac{1}{2} \frac{M_1}{\alpha E}$	$\frac{5}{4} \frac{M_1 + M_1^{\text{hf}}}{\beta E}$	$\frac{3}{4} \frac{M_1 - M_1^{\text{hf}}}{\beta E}$

polarization $\mathbf{P}^{(2)}$ available in $\Delta F = 0$ hfs components. This polarization is induced along the beam by interference between the second and third terms of (2) ($\alpha E \cdot \beta E$ interference) when the beam polarization is made circular. When a dc magnetic field is temporarily applied along y (Fig. 1a), $\mathbf{P}^{(2)}$ acquires by Hanle effect a non-zero component along x . This component is detected with the same optics and detector as $\mathbf{P}^{(1)}$. It is even under reversal of $\mathbf{E}$. In the $\Delta F = 0$ case, we obtain $P^{(1)}/P^{(2)}$ after completion of successive measurements of $P^{(1)}$ in zero magnetic field and $P^{(2)}$ in the Hanle field. In a known Stark field E , this ratio yields M'_1/β , i.e. M_1/β according to (1). On the other hand, the ratio of the polarizations $P^{(1)}$ measured in the $3 \rightarrow 4$ and $4 \rightarrow 3$ components directly yields M_1^{hf}/M_1 . A great advantage of the Stark interference method is to detect without any background from Cs-Cs collisions or Cs_2 dimers. The reason is that $\mathbf{P}^{(1)}$ is created along $\hat{k} \times \mathbf{E}$, which averages to zero when the Stark field $\mathbf{E}$ is replaced by a collisional or intramolecular field of random direction. This argument does not apply to $\mathbf{P}^{(2)}$, created in a direction independent of $\mathbf{E}$ (the direction of the beam), so that a background can exist for $\mathbf{P}^{(2)}$. Yet from auxiliary measurements it turns out to be negligible ($\ll 10^{-3}$) in our experimental conditions.

The set-up is schemed in figure 1b. The x component of the 7S state polarization is detected through the circular polarization ratio of the $7S_{1/2}$ - $6P_{1/2}$ fluorescence light in the x direction. A polarization modulator labels the circular and linear components of the laser beam polarization with characteristic frequencies and phases [12], thus enabling further distinction between $P^{(2)}$ and $P^{(1)}$ by lock-in detection. The possible calibration difference between the two electronic channels of detection is eliminated in either of two ways :

i) Half of the time, a $\lambda/4$ plate is inserted after the polarization modulator, with its axes along the bissectrices of xy . This interchanges the specific modulations of $P^{(1)}$ (linear polarization along xy) and $P^{(2)}$ (circular polarization). Each channel thus detects $P^{(1)}$ and $P^{(2)}$ in turn. Measurements of the ratio $P^{(1)}/P^{(2)}$ with and without the plate are then combined.

ii) The $\lambda/4$ plate is permanently inserted, with its axes along xy . This leaves the modulation of $P^{(1)}$ unchanged, but brings that of $P^{(2)}$ to the same frequency, in phase quadrature with the former. $P^{(1)}$ and $P^{(2)}$ are then detected in one and the same two-phase lock-in amplifier.

The polarization $P^{(1)}$ is odd under laser beam reversal. It is therefore important to avoid spurious reflection of the beam back to the observation region. This is achieved by reflecting the beam from a tilted mirror onto a stop inside the Cs cell. For better discrimination of $P^{(1)}$, the Stark field is (randomly) reversed and only odd signal is retained. The details of the apparatus are the same as in reference [8], except the tilted mirror ensuring *single pass* of the beam.

Systematic uncertainty in the measurement of M_1/β mainly originates in electronics calibration ($\sim 0.6\%$ from lock-in sensitivity change between $P^{(1)}$ and $P^{(2)}$) and in uncertainty in the value of the electric field ($\sim 0.8\%$ due to the finite size and imperfect parallelism of the capacitor plates, and to wall effects). Other possible imperfections (in the polarization modulator, $\lambda/4$ plate retardation and orientation, E reversal, fluorescence polarization analyser, drifts) were found to have smaller effects. In the M_1^{hf}/M_1 value, extracted from measurements of $P^{(1)}$ in the $4 \rightarrow 3$ and $3 \rightarrow 4$ hfs components, a correction of 2.8% was included to account for a small contribution of the wing of the strong adjoining $\Delta F = 0$ component. The uncertainty concerning this correction is the main systematic uncertainty.

Results. — We first checked (1) by comparing $P^{(1)}$ on the various hfs components : i) the ratio $P^{(1)}(3 \rightarrow 3)/P^{(1)}(4 \rightarrow 4)$ agrees with the expected $3/5$ value (cf. table I) at the percent level ; ii) the ratios $M_1'(3 \rightarrow 4)/M_1'(4 \rightarrow 3)$ and $M_1'(3 \rightarrow 4)/M_1'(4 \rightarrow 4)$ give two independent determinations of M_1^{hf}/M_1 which agree within statistical accuracy ($\sim 8\%$). The expected $1/E$ dependence of the ratio $P^{(1)}/P^{(2)}$ was then checked in the $4 \rightarrow 4$ component between 300 and 1 000 V/cm (Fig. 2). From measurements using the $\lambda/4$ plate in either of the two ways described above, we respectively find :

$$M_1/\beta = -29.40 \pm 0.04 \text{ V/cm} \quad \text{and} \quad -29.41 \pm 0.05 \text{ V/cm}$$

(where the quoted uncertainty is only statistical). The agreement between these results is an indication that instrumental calibration is eliminated correctly. When combined, they give :

$$M_1/\beta = -29.40 \pm 0.03 \text{ (stat.)} \pm 0.44 \text{ (sys.) V/cm} . \quad (3)$$

As a cross-check, measurements performed in a different cell (with more rigorous capacitor plate parallelism) and using a more accurate lock-in amplifier, yield :

$$M_1/\beta = -29.71 \pm 0.17 \text{ (stat.)} \pm 0.30 \text{ (sys.) V/cm} . \quad (4)$$

Combining values (3) and (4), we finally obtain :

$$M_1/\beta = -29.55 \pm 0.45 \text{ V/cm} . \quad (5)$$

Fig. 2. — Ratio $P^{(1)}/P^{(2)}$ versus inverse of electric field; rms statistical uncertainty on each point is 1 %.

The value of M_1^{hf}/M_1 was extracted from 15 independent measurements of the ratio $M_1'(3 \rightarrow 4)/M_1'(4 \rightarrow 3)$. Each time M_1' was separately measured in both hfs transitions, sometimes in one order, sometimes in the reversed order, to eliminate possible drifts of the Cs vapour pressure. The final value is :

$$M_1^{\text{hf}}/M_1 = 0.185 \pm 0.008 \text{ (mainly stat.)} . \quad (6)$$

The positive sign means that the modulus of the M_1' amplitude is larger for the 6S, $F = 3 \rightarrow 7S$, $F = 4$ component than for the 6S, $F = 4 \rightarrow 7S$, $F = 3$ component. Combining (5) and (6), we obtain :

$$M_1^{\text{hf}}/\beta = -5.47 \pm 0.31 \text{ V/cm} \quad (7)$$

as indicated earlier in reference [13].

To extract from (5) and (7) absolute values of M_1 and M_1^{hf} a value of β is needed. From the existing semi-empirical values [10] of α combined with the measured value [11] (-9.9 ± 0.1) of α/β , we adopt $\beta = (26.3 \pm 0.8) a_0^3$ as a reasonable average which agrees within a few percent with the theoretical value [9]. This leads to :

$$M_1 = -(4.13 \pm 0.18) \times 10^{-5} |\mu_B/c| , \quad (8a)$$

$$M_1^{\text{hf}} = -(0.763 \pm 0.066) \times 10^{-5} |\mu_B/c| . \quad (8b)$$

where we have added linearly the uncertainty in β to the experimental uncertainties involved in (5) and (7).

Discussion. — Seven years ago, we obtained the first experimental determination of M_1 [6], using the same basic method but fairly primitive apparatus and different techniques : instead of the present clean polarization modulation, we then used static polarizations and a modulated E field. In this old experiment, we overlooked the effect of the light reflected back by the exit

window which leads to a reduction of the $\alpha E. M_1$ interference signal and an increase of the $\alpha E. \beta E$ one, each of about $(10 \pm 3) \%$. Applying the appropriate correction to our old result, $M_1/\beta = -23.2 \pm 1.3$, we obtain $M_1/\beta = -27.8 \pm 2.7$. So the discrepancy with respect to our more accurate new result -29.55 ± 0.45 practically vanishes. At that time also, use of somewhat inaccurate values of α and α/β to extract M_1 happened to compensate the imperfect M_1/β nearly exactly. This explains why our new result for M_1 nearly coincides with our previous result. This M_1 value (Eq. (8a)) is to be compared to the theoretical approximate prediction [5] : $M_1 = -6.3 \times 10^{-5} |\mu_B/c|$.

Concerning now M_1^{hf}/M_1 , our result (Eq. (6)) is in conflict with the value measured by Hoffnagle *et al.* [7] who quote the results of three independent determinations : 0.36(5), 0.29(3) and 0.36(12). All of them involve the detection of an M_1^2 term in the fluorescence intensity in the presence of a non-negligible background. As discussed below, it turns out that our result leads to a value of M_1^{hf} in agreement with the theoretically predicted value while the above ones lead to serious discrepancy.

The matrix element of the magnetic dipole μ between the $|nS_{1/2} F\rangle$ and $|n' S_{1/2} F'\rangle$ states perturbed to first order by the hyperfine interaction [2] can be written :

$$\langle n' F' m' | \mu | n F m \rangle = \frac{-|\mu_B|}{E_{n'} - E_n} [\langle n' F' | H^{\text{hf}} | n F' \rangle - \langle n' F | H^{\text{hf}} | n F \rangle] \langle F' m' | \sigma | F m \rangle. \quad (9)$$

If the matrix elements $H_{n'FnF}^{\text{hf}}$ are calculated within the two following approximations : i) single-particle description of the $nS_{1/2}$ states; ii) non-relativistic approximation for the one-particle wave function, then we obtain the simple result :

$$H_{n'FnF}^{\text{hf}} = A(F) R_n(0) R_{n'}(0),$$

valid whether $n' = n$ or $n' \neq n$. $R_n(0)$ represents the radial $nS_{1/2}$ wave function at the origin and $A(F) = (4/3) \mu_0 \mu_B g_N \mu_N \langle F | \mathbf{S} \cdot \mathbf{I} | F \rangle$ does not depend on the principal quantum number. It is then clear that the off-diagonal element obeys the simple factorization relation :

$$|H_{n'FnF}^{\text{hf}}|^2 = H_{n'Fn'F}^{\text{hf}} H_{nFnF}^{\text{hf}} \quad (10)$$

which combined with (9) and the definition of M_1^{hf} leads to the prediction :

$$M_1^{\text{hf}} = -|\mu_B/c| \frac{[\Delta W_{6S}^{\text{hf}} \Delta W_{7S}^{\text{hf}}]^{1/2}}{E_{7S} - E_{6S}} = -0.806 \times 10^{-5} |\mu_B/c| \quad (11)$$

involving only the hf splittings of the $7S_{1/2}$ and $6S_{1/2}$ states [14] and their energy difference.

If, now, Dirac wave functions are used, the factorization does not hold exactly. However, it is possible to show, by integration by parts of the integral involving the radial Dirac wave functions, that the matrix elements of H^{hf} in the case of heavy elements receives its dominant contribution from values of the radial coordinate $r \lesssim a_0/Z$ [15]. In this region where the potential energy is for valence states much larger than the binding energies, the radial wave functions of the $nS_{1/2}$ states are found to coincide almost exactly, up to a normalization factor. It follows that the factorization relation (10) still holds in the single-particle model with relativistic wave functions (2).

(2) In reference [15] the hfs splittings of the $nS_{1/2}$ states for $6 \leq n \leq 10$ are calculated in the single-particle approximation with relativistic effects and reproduce the experimental results within a few per cent.

The many-body effects associated with the magnetic polarization of the core induce an effective *non-local* hyperfine interaction which involves exchange integrals between the $nS_{1/2}$ valence wave functions and the core wave functions. In the core region, the valence S wave functions are no longer proportional to one another. The factorization relation (10) is likely to be broken. For this reason, any deviation from the value of M_1^{hf} predicted by (11) would be a sign of presence of many-body effects. A serious discrepancy would cast some doubt on the validity for atomic caesium of the single-particle approximation for evaluating effects associated with short-range electron-nucleus interactions, like for instance the parity violating interaction. From the agreement at the level of 10 % between the experimental and predicted values of M_1^{hf} (Eqs. (8b) and (11)), we conclude that many-body effects which violate the factorization relation do not exceed 10 %.

Note added in proof. — After the manuscript was submitted we were informed by C. E. Wieman of an independent measurement by S. L. Gilbert, R. N. Watts and C. E. Wieman. Their result, $M_1 = -(4.21 \pm 0.08) \times 10^{-5} \mu_B/c$ and $M_1^{\text{hf}} = (0.759 \pm 0.055) \times 10^{-5} \mu_B/c$, agrees with ours. The opposite sign for M_1^{hf} comes from a different definition.

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