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Observation of the 6S1/2-7S1/2 single-photon transition of cesium induced by an external d. c. electric field — Source link \square

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OBSERVATION OF THE $6S_{1/2}$ - $7S_{1/2}$ SINGLE-PHOTON TRANSITION OF CESIUM INDUCED BY AN EXTERNAL d. c. ELECTRIC FIELD

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Résumé. — Nous avons observé l'excitation dipolaire électrique à un photon de l'état 7 ${}^{2}S_{1/2}$ du césium en excitant directement à l'aide d'un laser à colorant accordable continu, monomode, l'état fondamental 6 ${}^{2}S_{1/2}$ dans un champ électrique de 10 à 1 000 V/cm. La résonance est détectée en observant la fluorescence émise à partir de l'état excité. Nous avons vérifié les règles de sélection des transitions hyperfines, qui dépendent de l'orientation relative de la polarisation linéaire du faisceau et du champ électrique. En polarisation normale, le spectre révèle l'existence d'une partie de la polarisabilité dépendant du spin, induite par effet spin-orbite, dont nous avons pu déduire la grandeur par rapport à la partie scalaire. Les résultats en champ électrique faible montrent que la force d'oscillateur de la transition 6S-7S est inférieure à 2 $\times 10^{-13}$.

Abstract. — We have observed the direct single-photon electric dipole excitation of the 7 ${}^{2}S_{1/2}$ state of cesium, by exciting the 6 ${}^{2}S_{1/2}$ ground state in an external d. c. electric field of 10 to 1 000 V/cm with a single-mode, c. w., tunable dye laser. The resonance was detected by monitoring the decay fluorescence from the excited state. We verified the selection rules obeyed by the hyperfine transitions which depend on the relative orientation of the linear polarization of the beam with respect to the field. When they are orthogonal, the line pattern reflects the existence of a spin-dependent part of the polarizability induced by spin-orbit effects, the size of which was measured with respect to that of the scalar part. Results in very weak fields show that the oscillator strength of the 6S-7S transition is smaller than 2 \times 10⁻¹³.

We have detected the hyperfine structure of the $6S_{1/2}$ - $7S_{1/2}$ radiative transition of neutral cesium atoms placed in an external d. c. electric field using single-photon electric dipole excitation from the $6S_{1/2}$ ground state by a single-mode c. w. tunable dye laser. In the conditions of our experiment the transition probability was smaller by 8 to 12 orders of magnitude than that of the resonance line. In spite of the Doppler broadening the linewidth (0.75 GHz in our experiment) was sufficiently small to resolve fully the excitedstate and ground-state hyperfine structures. We were able to verify the different selection rules obeyed by the transition versus the angle between the applied electric field and the linear polarization of the exciting beam. We believe this is the first observation of a direct excitation from an alkali $S_{1/2}$ ground-state to the first-excited $S_{1/2}$ state and of the typical selection rules for electric dipole transitions induced by an electric field between hyperfine levels of two S states. Our measurements also yield a stringent upper limit to the transition rate in zero field.

High members ($n \approx 15-30$) of the forbidden series $n_0 S_{1/2} \rightarrow nS_{1/2}$ have been observed in the absorption

spectrum of K, Rb and Cs in electric fields of 300 to 1 000 V/cm [1]. For low quantum numbers the mixing of states of different l is much less effective, and direct excitation using conventional sources does not seem feasible. Recent theoretical investigations joint to the new possibilities offered in spectroscopy by tunable lasers have given a renewal of interest to those transitions. In a search for parity violating effects induced in atomic physics by weak neutral currents, particular attention was paid to twice forbidden magnetic dipole transitions $nS_{1/2}$ -n' $S_{1/2}$ of heavy alkali atoms [2].

The present paper, although devoted to effects induced by an external electric field, can be considered as a preliminary step into the rather difficult experimental investigation that consists in probing parity conservation in the excitation of a $nS_{1/2}$ - $n'S_{1/2}$ radiative transition.

Our experiment was performed on Cs vapor at high density $(10^{-2} \text{ to } 10^{-1} \text{ torr})$, contained in a glass cell without buffer gas; the cell was equipped with two plane parallel electrodes made of stainless steel and distant of 0.5 cm. The d. c. voltage applied could reach at least 500 V without discharge in the cell. The tunable light source was a Coherent Radiation model 490 jet-stream dye laser using fluorescein added

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with cyclooctatetraene and pumped by a Coherent Radiation model CR8 argon ion laser. A piezo-driven, thermally stabilized, air-spaced Fabry-Pérot etalon placed inside the main cavity insured single-mode

Radiation model CR8 argon ion laser. A piezo-driven, thermally stabilized, air-spaced Fabry-Pérot etalon placed inside the main cavity insured single-mode operation. The laser output mirror was mounted on a piezo-drive. Our measurements were performed with 50 mW single-mode laser power. The laser was tuned at 5 393 Å. The laser beam, chopped at 130 Hz, was directed into the cell at right angle with the electric field E_0 . Decay fluorescence perpendicular to E_0 and to the beam was collected by an f/1 lens doublet and imaged on to a germanium photodiode. An interference filter was introduced to cut down background light. The filter had a 720 Å bandwidth and passed light from only the $7S_{1/2}$ - $6P_{1/2}$ step in the decay from the $7S_{1/2}$ state, with a characteristic wavelength of 13 588 Å.

Measurements of the detailed structure of the $6S_{1/2}$ - $7S_{1/2}$ transition were made by shifting the laser frequency by successive amounts of the order of 300 MHz which were accurately measured with a confocal spherical Fabry-Pérot interferometer having a free spectral range of 1.5 GHz and a finesse of 100. The amplified modulated output from the photodiode was detected in a phase-sensitive lock-in amplifier. The laser light intensity was simultaneously monitored using the modulated output of a silicium photodiode. The ratio of both signals, i. e. the fluorescence rate, was plotted versus the laser frequency for an increasing set and a decreasing set of discrete values. A typical spectrum is shown in figure 1a. The largest peak corresponds to a fluorescence signal of 4×10^8 photons/s. The linear polarization of the incident beam (hereafter denoted by the unit vector $\boldsymbol{\varepsilon}$) had to be rotated by an arbitrary angle θ with respect to the field E_0 . The beam delivered by the laser was linearly polarized paralled to E_0 . We introduced before the cell a half-wave plate with axis at $\theta/2$ and a Glan prism with axis at θ . In this way the polarization of the incident beam was defined to better than 1 part in 10⁴ and the rotation of the polarization did not produce any noticeable loss in intensity.

Considerable changes in the spectrum intensity and structure were observed when θ was varied, as can be seen while comparing figure 1a ($\epsilon \parallel E_0$) and figure 1b $(\varepsilon \perp E_0)$. Note the change of scale : the fluorescence rate is smaller by 2 orders of magnitude in the second case. The four components observed in figure 1b are attributed to the four transition $6S_{1/2}(F) \rightarrow 7S_{1/2}(\mathcal{F})$ with the possible values 3 and 4 for the quantum number of the total angular momentum of cesium in the ground state (F) and excited state (F). Their spacings are in excellent agreement with the known hyperfine splittings, namely 9 192.63 MHz for the $6S_{1/2}$ state [3] and 2 175 MHz for the $7S_{1/2}$ state [4]. In figure 1a only the two components relative to the $F = \mathcal{F}$ transitions are observed. The amplitudes of all peaks are proportional to E_0^2 .

Interpretation of all above facts can be found in the



Frequency



7.018 GHz

5

rate /arb.

Fluorescence

1500

1000

500



Fig. 1 b.

FIG. 1. — Fluorescence rate (arbitrary units) versus incident laser beam frequency. Electric field 1 000 V/cm. Solid line : theoretical spectrum ; crosses : experimental points : (a) $\varepsilon \not/\!/ E_0$, (b) $\varepsilon \perp E_0$.

symmetry properties of the polarizability second rank tensor \mathbf{T} which relates the induced electric dipole \mathbf{D}_{eff} to the external field [5]:

$$\mathbf{D}_{\rm eff} = \mathbf{T}\mathbf{E}_0 \,,$$

and can be treated according to first order perturbation theory :

<

$$7S_{1/2} | D_{eff} | 6S_{1/2} > =$$

$$= < 7S_{1/2} | d.E_0 \frac{1}{E_{7S} - \Im C} d +$$

$$+ d \frac{1}{E_{6S} - \Im C} d.E_0 | 6S_{1/2} > , \quad (1)$$

where \mathcal{K} is the unperturbed atomic hamiltonian.

The following decomposition is implied by symmetry considerations :

$$<7S_{1/2} \sigma' | \mathbf{D}_{eff} | 6S_{1/2} \sigma > =$$
$$= \alpha_{7,6} \mathbf{E}_0 \, \delta_{\sigma'\sigma} + i\beta_{7,6} (\mathbf{\sigma} \wedge \mathbf{E}_0)_{\sigma'\sigma}, \quad (2)$$

coefficients explicitly written in reference [5] in terms of energies and radial wave functions of intermediate p-states. Besides its scalar part $\alpha_{7,6}$, the polarizability thus exhibits a vectorial part which is spin-dependent as a result of the spin-orbit coupling in the p-states admixed with the s-states. Note that the coefficient $\beta_{7,6}$ in eq. (2) is zero if spin-orbit coupling is not taken into account, since \mathcal{H} then commutes with σ . Thus $\beta_{7,6}$ is expected to be one order of magnitude less than $\alpha_{7,6}$ (¹).

With an incident beam of polarization ε , the $nS\sigma \rightarrow n'S\sigma'$ transition matrix element is proportional to :

$$<7S_{1/2} \sigma' | \mathbf{D}_{eff} \cdot \boldsymbol{\varepsilon} | 6S_{1/2} \sigma > =$$
$$= \alpha_{7,6} \mathbf{E}_0 \cdot \boldsymbol{\varepsilon} \delta_{\sigma\sigma'} + i\beta_{7,6} (\boldsymbol{\sigma} \wedge \mathbf{E}_0)_{\sigma'\sigma} \cdot \boldsymbol{\varepsilon} .$$
(3)

When \mathbf{E}_0 and $\boldsymbol{\varepsilon}$ are parallel, the effective dipole operator acts as a scalar on the atomic wave function, therefore only transitions conserving the total angular momentum are allowed, i. e. $\mathbf{F} = \mathcal{F}$; while, if \mathbf{E}_0 and $\boldsymbol{\varepsilon}$ are orthogonal, \mathbf{D}_{eff} acts as a spin-component and all transitions get allowed.

If the electronic spin is coupled with a nuclear spin 7/2 as in Cs, the following intensity ratios are then predicted for the different $6S_{1/2}$, $F \rightarrow 7S_{1/2}$, \mathcal{F} transitions (noted $F \rightarrow \mathcal{F}$ for abbreviation) :

$$4 \rightarrow 4 \qquad (36 \alpha^2 \cos^2 \theta + 15 \beta^2 \sin^2 \theta) E_0^2 \quad (4a)$$

$$3 \rightarrow 3 \qquad (28 \alpha^2 \cos^2 \theta + 7 \beta^2 \sin^2 \theta) E_0^2 \quad (4b)$$

 $3 \rightarrow 4 \text{ and } 4 \rightarrow 3$ $(21 \beta^2 \sin^2 \theta) E_0^2$ (4c)

(For the sake of simplicity, the indices 7,6 of α and β are now omitted.)

We have compared our experimental results with the theoretical spectrum which consists of 4 Doppler lines having the above intensity ratios, separations deduced from the known hyperfine splittings of the two S states :

$$\begin{bmatrix} v(4 \to 4) - v(4 \to 3) = \Delta W(7S_{1/2}); \\ v(4 \to 4) - v(3 \to 4) = \Delta W(6S_{1/2}) \end{bmatrix}$$

and a half-height linewidth of 0.750 GHz, the Doppler width for ¹³³Cs at T = 472 K. As can be seen from figure 1, the agreement with the experimental points is quite good. From the experimental ratio of fluorescence rates measured with $\varepsilon \not|/ \mathbf{E}_0$ and $\varepsilon \perp \mathbf{E}_0$ we can deduce $|\alpha/\beta|$.

We have also measured the dependene of the amplitudes of the peaks corresponding to the $4 \rightarrow 4$ and $3 \rightarrow 3$ transitions versus the angle θ between the d. c. field \mathbf{E}_0 and the polarization ε of the exciting beam. Results are shown in figure 2*a* and 2*b* and compared to the theoretical predictions of eq. (4*a*) and

(4b); the agreement is excellent and yields two other determinations of $|\alpha/\beta|$. Finally we can conclude that :

$$|\alpha/\beta| = 8.8 \pm 0.4$$

This value has the order of magnitude expected. The theoretical value [5] 7 should be considered only as a rough estimate in view of the large compensations arising between terms in the summation which gives β .



FIG. 2 b.

FIG. 2. — Peak intensity of the fluorescence rate (arbitrary units) versus the angle θ . Solid line : theoretical curve ; crosses : experimental points : (a) $6S_{1/2}$ (F = 4) $\rightarrow 7S_{1/2}$ (F = 4) transition ; (b) $6S_{1/2}$ (F = 3) $\rightarrow 7S_{1/2}$ (F = 3). The background is 4 units.

It is worth noting that our experiment presents some analogy with a two-photon excitation of an alkali $nS_{1/2} \rightarrow n'S_{1/2}$ transition; in both cases we expect the same transition pattern. However a difference arises for two-photon experiments without Doppler broadening [6] where the two photons have the same polarization and the same frequency : the polarizability tensor

 $^(^{1})$ The spin-dependent polarizability is closely related to the deviation from a 2 : 1 ratio observed for the intensities of the principal series doublet lines.

then becomes symmetric and cannot have any spindependent component, so that no $\mathcal{F} \neq F$ transition is allowed, even in presence of strong spin-orbit coupling.

Our measurements reported in figures 1 and 2 were performed in a d. c. electric field of 1 000 V/cm. In an attempt to observe the transition in zero field, we have followed the quadratic field-dependence of the $4 \rightarrow 4$ transition peak, in the configuration $\boldsymbol{\epsilon} /\!\!/ \mathbf{E}_0$, at much lower field amplitudes, namely in the range 0 to 40 V/cm. We concluded that the $4 \rightarrow 4$ transition rate in zero field — which we have not yet observed — is smaller than the one induced by an external field of 12 V/cm. Using the theoretical value of the induced electric dipole $\alpha_{7.6} E_0$ given in reference [5], we conclude that the oscillator strength of the forbidden transition in zero field is smaller than 2×10^{-13} . We can interpret this preliminary result in terms of an experimental upper limit to the magnetic dipole of the $6S_{1/2} \rightarrow 7S_{1/2}$ transition :

$$|M_1| \lesssim 3 \times 10^{-4} \mu_{\rm B}/c$$

- KUHN, H. G., Z. Phys. 61 (1930) 805.
 JENKINS, F. A. and SEGRE, E., Phys. Rev. 55 (1939) 545.
 NY TSI-ZE et CHOONG SHIN-PIAW, J. Physique 6 (1935) 147.
- [2] BOUCHIAT, M. A. and BOUCHIAT, C., J. Physique 35 (1974) 899.
- [3] ARDITI, M. and CARVER, T. R., Phys. Rev. 109 (1958) 1012 and Phys. Lett. 48B (1974) 111.
- [4] GUPTA, R., HAPPER, W., LAM, L. K. and SVANBERG, S., Phys. Rev. 8A (1973) 2792.

 $\mu_{\rm B}$ being the Bohr magneton. We can also deduce an upper limit to the electric dipole of transition induced by parity violation :

$$|E_1| \leq 10^{-6} e a_0$$
 (in atomic units).

We recall that $|M_1|$ is theoretically estimated to be of the order of $10^{-4} \mu_B/c$ [2], thus we believe that we shall be able to detect it in forthcoming measurements. At the same time, improvement of the quite preliminary limit given to $|E_1|$ is to be expected, in particular by measuring not only the square of the amplitude but also interference effects which are much more sensitive to a small amplitude. Although the present limit is rather large as compared with the theoretical estimate for the electric dipole of transition induced by weak neutral currents [2], namely larger by a factor 10^5 , however it is already more significative than the one deduced by Bradley and Wall [7] from their experiment on molecular oxygen, which represents 5×10^6 of the theoretical estimate in this case.

References

- [5] BOUCHIAT, M. A. and BOUCHIAT, C., J. Physique 36 (1975) 493.
- [6] BIRABEN, F., CAGNAC, B. and GRYNBERG, G., Phys. Rev. Lett. 32 (1974) 643.
 - LEVENSON, M. D. and BLOEMBERGEN, N., Phys. Rev. Lett. 32 (1974) 645.
- [7] BRADLEY III, L. C. and WALL, N. S., Nuovo Cimento 25 (1962) 48.