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OBSERVATION OF THE INTERFERENCE BETWEEN THE MAGNETIC AND ELECTRIC DIPOLE AMPLITUDES OF THE CAESIUM 6S-7S TRANSITION IN AN ELECTRIC FIELD. MEASUREMENT OF THE TRANSITION MAGNETIC MOMENT

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Résumé. — Nous avons observé et mesuré le moment magnétique d'une transition S-S très fortement interdite d'un atome alcalin. En réalisant l'excitation à un seul photon de la transition $6S_{1/2}$-$7S_{1/2}$ du césium dans un champ électrique quasi-statique, nous avons pu détecter une polarisation électronique de l'état excité, qui est spécifique d'un effet d'interférence susceptible de se produire dans les transitions dipolaires mixtes, à la fois électriques et magnétiques. Nos mesures, indépendantes de la pression de vapeur, indiquent que l'amplitude dipolaire magnétique M_1 est égale en module à l'amplitude dipolaire électrique E_1, induite par un champ électrique parallèle à la polarisation de l'onde incidente et d'amplitude 2,62 ± 0,21 V/cm. Nous obtenons aussi le signe relatif de M_1 et E_1. En utilisant une valeur théorique de E_1 et une convention de phase définie, nous en déduisons le moment magnétique de transition

$$\langle 7S, \tfrac{1}{2} \mid \mu_z \mid 6S, \tfrac{1}{2} \rangle = -(4{,}24 \pm 0{,}34)\, 10^{-5} \mid \mu_B \mid ,$$

en bon accord avec une estimation théorique. Ce résultat s'exprime aussi sous forme de force d'oscillateur en champ électrique nul : $f_{6S\text{-}7S} = 4{,}05 \times 10^{-15}$, ou de probabilité d'émission à un photon (M_1) $\Gamma_{7S\text{-}6S} = 0{,}93 \times 10^{-6}\ s^{-1}$.

Abstract. — We report here the first observation and measurement of the transition magnetic moment for a strongly forbidden S-S transition of an alkali atom. By using single-photon excitation of the $6S_{1/2}$-$7S_{1/2}$ caesium transition in a quasi-static electric field, we have detected an electronic polarization of the excited state, specific to an interference effect occurring in mixed electric magnetic dipole transitions. Our measurements, independent of the vapour density, indicate that the magnetic dipole amplitude M_1 has the same size as the electric dipole amplitude E_1, induced by an electric field, parallel to the light wave polarization, and equal to 2.62 ± 0.21 V/cm. We also obtain the relative sign of M_1 and E_1. Using a theoretical value of E_1 and a well-defined phase convention, we deduce for the magnetic moment of the transition

$$\langle 7S, \tfrac{1}{2} \mid \mu_z \mid 6S, \tfrac{1}{2} \rangle = -(4.24 \pm 0.34)\, 10^{-5} \mid \mu_B \mid ,$$

in good agreement with a theoretical estimate. This result can also be expressed in terms of the oscillator strength in zero electric field $f_{6S\text{-}7S} = 4.05 \times 10^{-15}$, or of the single-photon M_1 decay rate $\Gamma_{7S\text{-}6S} = 0.93 \times 10^{-6}\ s^{-1}$.

We have investigated experimentally the influence of an external static electric field on a radiative single-photon S-S transition by focussing our attention on the interference effect resulting from the simultaneous existence of two contributing transition amplitudes, the induced electric dipole (E_1) and the magnetic dipole (M_1). We report here on the observation of the $E_1 M_1$ interference, a new effect which provides us with the first determination of M_1 in size and sign for a strongly forbidden $S_{1/2}$-$S_{1/2}$ alkali transition.

Our result for caesium,

$$\langle 7S \mid \mu_z \mid 6S \rangle = -4.24 \times 10^{-5} \mid \mu_B \mid ,$$

indicates that the 6S-7S transition is nearly as strongly forbidden as the still unobserved 1S-2S single-photon transition of atomic hydrogen, for which a theoretical value of $7.4 \times 10^{-6} \mid \mu_B \mid$ is predicted [1]. This was

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the order of magnitude expected for caesium. Orthogonality of the two S wave-functions suppresses any first-order effect; among the mechanisms which can give rise to a non-vanishing transition amplitude, it was natural to consider the relativistic corrections to the magnetic moment which are responsible for a deviation between the gyromagnetic ratio of the Cs ground state and that of the free electron observed experimentally [2],

$$\frac{\Delta g}{g} = \frac{g_{Cs} - g_e}{g_e} \simeq 1.1 \times 10^{-4} .$$

The correct qualitative interpretation for a heavy atom like caesium requires the combined effect of configuration mixing and of spin-orbit coupling [3]. This effect also produces a transition magnetic moment which, expressed in units of μ_B, was shown to be of the same order of magnitude as $\Delta g/g$, but reduced by a factor of the order of $\frac{1}{2}$ to $\frac{1}{3}$ [4]. Furthermore the measurement of M_1 can be considered to be an important step in the attempt to look for parity violation in a strongly forbidden transition [5].

The interference effect observed in our experiment is typical of a mixed $E_1 M_1$ transition. As shown theoretically [5], when atoms in a d.c. electric field are excited through a $S_{1/2}$-$S_{1/2}$ transition by a resonant beam of direction $\hat{\mathbf{k}}_i$, the $E_1 M_1$ interference gives rise to an atomic orientation in the upper state parallel to $\hat{\mathbf{k}}_i \wedge \mathbf{E}_0$. To obtain physical insight into this effect, let us consider the emission process by using a classical picture. The radiating atom is described by two dipoles, an electric one along $\mathbf{E}_0$ and a magnetic one along the electron spin $\mathbf{S} = \boldsymbol{\sigma}/2$, both oscillating in phase at the optical frequency ω :

$$\mathbf{d} = \mathbf{D}\, e^{-i\omega t} = E_1\, \hat{\mathbf{E}}_0\, e^{-i\omega t} \qquad (1a)$$

($\hat{\mathbf{E}}_0$ unit vector along $\mathbf{E}_0$),

$$\mathbf{m} = \frac{\boldsymbol{\mu}}{c} e^{-i\omega t} = M_1\, \boldsymbol{\sigma}\, e^{-i\omega t} . \qquad (1b)$$

The electric field and magnetic induction radiated are well known in classical electrodynamics. The average power radiated per unit solid angle is :

$$\frac{d\mathcal{P}}{d\Omega} = \frac{c}{8\pi} k^4 \left[|\, \mathbf{D} \wedge \hat{\mathbf{k}}\, |^2 + \left| \frac{\boldsymbol{\mu}}{c} \wedge \hat{\mathbf{k}} \right|^2 + 2\, \frac{\boldsymbol{\mu}}{c} \cdot (\hat{\mathbf{k}} \wedge \mathbf{D}) \right] . \qquad (2)$$

Three contributions appear in the above expression : the first two represent the power radiated by the two dipoles oscillating each one alone, the third results from interference between the fields radiated by the two sources. It is positive and maximum when the relative orientation and magnitude of the two dipoles satisfy the condition $\boldsymbol{\mu}/c = \hat{\mathbf{k}} \wedge \mathbf{D}$; the radiated power is then doubled. Conversely when

$$\boldsymbol{\mu}/c = - \hat{\mathbf{k}} \wedge \mathbf{D}$$

the interference term can exactly cancel the sum of the two direct terms. Thus, if the atomic spin is oriented in the excited state along $\hat{\mathbf{k}} \wedge \mathbf{E}_0$, its emission probability is enhanced in one of the two directions $+ \hat{\mathbf{k}}$ or $- \hat{\mathbf{k}}$ — depending on the relative sign of E_1 and M_1 — and quenched in the opposite direction. Since a similar result holds for the absorption probability, we expect the excitation by a plane wave of momentum $\mathbf{k}_i$, to produce a certain degree of spin orientation along $\hat{\mathbf{k}}_i \wedge \mathbf{E}_0$, equal to

$$2\, E_1\, M_1/(E_1^2 + M_1^2) .$$

It is important to note that the interference term is a linear function of the applied electric field, and is reversed when either $\mathbf{E}_0$ or $\hat{\mathbf{k}}_i$ is reversed. A quantum-mechanical derivation of this result [5], taking into account the nuclear spin I, predicts the following result for the electronic polarization $\mathbf{P}_e^{(1)} = 2 \langle \mathbf{S} \rangle$, when the incident polarization is linear, parallel to $\mathbf{E}_0$:

$$\mathbf{P}_e^{(1)} = \frac{8}{3} \frac{F(F+1)}{(2I+1)^2} \frac{M_1}{E_1} \left[1 + \frac{4F(F+1)}{3(2I+1)^2} \frac{M_1^2}{E_1^2} \right]^{-1} \times \hat{\mathbf{k}}_i \wedge \hat{\mathbf{E}}_0 ; \qquad (3)$$

$F = I \pm \frac{1}{2}$ is the total quantum number of the hyperfine levels involved in a $\Delta F = 0$ transition. M_1 is defined as :

$$M_1 = \left\langle 7S, m_S = \frac{1}{2} \left| \frac{\mu_z}{c} \right| 6S, m_S = \frac{1}{2} \right\rangle \qquad (4)$$

with $\mu_z = - |\, \mu_B \,|\, \sigma_z$ for the free electron. E_1 has been computed, using first-order perturbation theory [5], with conventions where the radial wave functions are all supposed positive near the origin. The result is :

$$E_1 = - |\, e \,| \langle 7S, m_S \,|\, z \,|\, 6S, m_S \rangle = 1.62 \times 10^{-5} \frac{|\, \mu_B \,|}{c} E_{0z}\ (\mathrm{V/cm}) . \qquad (5)$$

A theoretical computation of the Cs ground state static polarizability using the same ingredients reproduces the experimental value within an accuracy of 5 %, so we feel that the uncertainty in the theoretical value of E_1 should not exceed 10 to 15 %.

In our experiment, the atomic orientation $\mathbf{P}_e$ created in the excited state is monitored through the circular polarization of the decay fluorescence light corresponding to the allowed $7S_{1/2}$-$6P_{1/2}$ transition. The light intensity transmitted through a circular analyzer onto the detector reflects $\mathbf{P}_e$, according to :

$$\mathcal{L}_f(\xi_f) = \tfrac{1}{2}(1 + \xi_f\, \mathbf{P}_e . \hat{\mathbf{k}}_f)\, I_f ; \qquad (6)$$

I_f is the total intensity collected in the direction $\hat{\mathbf{k}}_f$,

ξ_f is the sense of rotation of the transmitted circular polarization

$$\left(\mathbf{e} = \frac{1}{\sqrt{2}}(\hat{\mathbf{x}} + i\xi_f \hat{\mathbf{y}}) \quad \text{with} \quad \hat{\mathbf{x}} \wedge \hat{\mathbf{y}} = \hat{\mathbf{k}}_f\right).$$

Experimentally we observe the quantity $P_e^{(1)} I_f$ in conditions such that $E_1 \gg M_1$. We then expect $P_e^{(1)}$ to go like E_1^{-1}, i.e. to be inversely proportional to E_0, while I_f, proportional to E_1^2, varies like E_0^2; thus $P_e^{(1)} I_f$ is proportional to E_0. As described below, we have actually been able to observe this; for $E_0 = 10^3$ V/cm, it indicates that $P_e^{(1)}$ is of the order of 1.5×10^{-3}.

Before obtaining M_1/E_1 from the measurement of $P_e^{(1)}$ it was necessary to correct for some depolarization of the excited state due to collisions with ground state atoms in the vapour. Imperfections of the optical system are another possible source of light depolarization. For an absolute measurement of M_1, it is best to avoid this depolarization correction, whatever its origins. We proceed by calibrating $P_e^{(1)}$ in terms of another polarization $P_e^{(2)}$ whose absolute value is known from an independent measurement. For $P_e^{(2)}$ we use the upper state polarization resulting from another interference effect which involves the two terms of the induced electric dipole matrix element :

$$\langle 7S_{1/2}, m_S' \mid \mathbf{D} \mid 6S_{1/2}, m_S \rangle = -\alpha \mathbf{E}_0 \delta_{m_S' m_S} - 2i\beta(\mathbf{S} \wedge \mathbf{E}_0)_{m_S' m_S}, \quad (7)$$

where m_S and m_S' denote the spin components of the lower and upper states and α and β are real coefficients describing the spin-independent and spin-dependent parts of the Cs polarizability for the 7S-6S transition [5]. Note that α/β has already been measured [6] :

$$|\alpha/\beta| = 8.8 \pm 0.4. \quad (8)$$

It is known, both theoretically and experimentally [7], that a circularly polarized light beam produces in the excited state an electronic polarization directed along the beam, having the modulus :

$$P_e^{(2)} = \frac{8}{3}\frac{F(F+1)}{(2I+1)^2}\left|\frac{\beta}{\alpha}\right|\left[1 + \frac{4F(F+1)}{3(2I+1)^2}\frac{\beta^2}{\alpha^2}\right]^{-1}; \quad (9)$$

as all the parameters are known this equation gives the absolute value of $P_e^{(2)}$. Note that the quantity $P_e^{(2)} I_f$ accessible to our experiment varies as E_0^2.

In our experiment, caesium atoms in a vapor (10^{-2} to 10^{-1} torr) are excited by a single-mode, c.w. laser beam, tuned at the resonant frequency of the 6S-7S transition ($\lambda = 5\,393.5$ Å), in an external field E_0 (500 to 1 000 V/cm) perpendicular to the beam [6]. Resonance is detected by monitoring the decay fluorescence on the $7S_{1/2}$-$6P_{1/2}$ transition, in the direction $\hat{\mathbf{k}}_f = \hat{\mathbf{k}}_i \wedge \hat{\mathbf{E}}_0$. To make the measurement of $P_e^{(1)}$ the incident light is linearly polarized along $\mathbf{E}_0$, so that the spin dependent term proportional to β in (7) does not contribute.

Our apparatus is schematized in figure 1. The fluorescence polarization is analyzed with a quarterwave plate RQ, rotating at frequency $\omega_P/2$, followed by a fixed polarizer P. The transmitted intensity thus presents a modulation $I_f P_e^{(1)} \cos \omega_P t$. The « d.c. » electric field is in fact modulated at a frequency

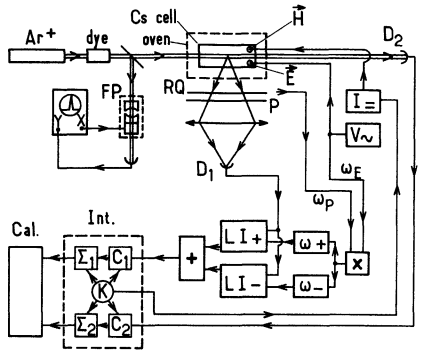

FIG. 1. — Schematic view of the apparatus. ω_+, ω_- : selective amplifiers ; C_1, C_2 : analog to digital converters ; Σ_1, Σ_2 : accumulators. Other symbols are defined in the text.

of 700 Hz ($E_0 \cos \omega_E t$). Since the quantity $I_f P_e^{(1)}$ is linear in the electric field, the transmitted intensity is modulated by the product $\cos \omega_E t \cos \omega_P t$. This modulation is detected by two lock-in amplifiers LI_+ and LI_- tuned to $\omega_+ = \omega_E + \omega_P$ and $\omega_- = \omega_E - \omega_P$. Their outputs are added. The result, as well as the laser intensity i, are fed into a 2-channel digital integrator, monitored by a clock K. After 8 seconds' integration, the two results are sent to the calculator for repeated accumulation. The signal-to-noise ratio is typically equal to 1 after 8 second-integration. The laser frequency, scanned with a Fabry-Pérot interferometer, is kept resonant to within 1/10 of the Doppler width during data accumulation. The ratio of the two signals gives the cross-modulated fluorescence yield $\mathcal{S}^{(1)}$:

$$\mathcal{S}^{(1)} = \kappa P_e^{(1)} I_f/i = \kappa P_e^{(1)} \mathcal{S}_t. \quad (10)$$

The factor κ accounts for all depolarization processes. The fluorescence yield $\mathcal{S}_t = I_f/i$ is obtained by separate measurement by modulation of the incident light intensity.

The signals $\mathcal{S}^{(1)}$ and $\mathcal{S}_t$ have a strong resonant behaviour with possibly a weak non resonant background. $\mathcal{S}^{(1)}$ is found, as expected, proportional to the electric field, within the statistical spread of our results. The ratio of measurements performed on the $4 \rightarrow 4$ and on the $3 \rightarrow 3$ transitions are also in quite good agreement with the predicted theoretical value of $\frac{5}{3}$. Finally, to confirm the interpretation of the signal $\mathcal{S}^{(1)}$ as coming

from an atomic orientation of the $7S_{1/2}$ state along $\hat{\mathbf{k}}_f$, we applied a d.c. magnetic field $\mathbf{H}$ along $\mathbf{E}_0$ and observed the lorentzian H-dependence of the signal (*Hanle effect*). The width $\Delta H \approx 9$ G is well accounted for by the lifetime of the 7S state [8] and a slight collision broadening, and agrees with a more precise determination to be given below. We have made use of this H-dependence to eliminate an eventual residual background on $\mathcal{S}^{(1)}$ by a sequential measurement of the difference

$$\mathcal{S}^{(1)}(H = 0) - \mathcal{S}^{(1)}(H \gg \Delta H) .$$

We now outline how the sign of E_1/M_1 is extracted from this experiment. The unit vector $\hat{\mathbf{E}}_0$ along $\mathbf{E}_0$ being defined in such a way that $\mathbf{E}_0 = | E_0 | \hat{\mathbf{E}}_0 \cos \omega_E t$, let $\varepsilon_1 = \hat{\mathbf{k}}_f . (\hat{\mathbf{k}}_i \wedge \hat{\mathbf{E}}_0)$. Using a luminescent photodiode, we make a reference light signal varying in time as $1 + \cos \omega_E t$, that is circularly polarized in the sense defined by ε_2 (i.e. with a polarization vector

$$\mathbf{e} = \frac{1}{\sqrt{2}} (\hat{\mathbf{x}} + i\varepsilon_2 \hat{\mathbf{y}}) ,$$

$\hat{\mathbf{x}} \wedge \hat{\mathbf{y}}$ being in the direction of propagation). Finally this circularly polarized light signal is detected exactly in the same way as the fluorescence light, which gives a value $\mathcal{S}^{(0)}$ to be compared with $\mathcal{S}^{(1)}$. Let $\varepsilon_3 =$ sign of $\mathcal{S}^{(0)}/\mathcal{S}^{(1)}$. From eq. (3) and (6), we conclude that sign $(E_1/M_1) = \varepsilon_1 \varepsilon_2 \varepsilon_3$. Experimentally we find :

$$E_1/M_1 < 0 , \qquad (11)$$

i.e. the angular electronic polarization $\mathbf{P}_e^{(1)}$ in a d.c. field $\mathbf{E}_0$ has the direction and sense of $\mathbf{E}_0 \wedge \hat{\mathbf{k}}_i$.

For the measurement of $P_e^{(2)}$, only a few straightforward modifications have to be applied to the preceding experiment (see Fig. 2) : 1) the polarization

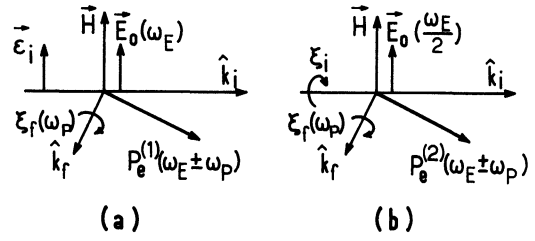

FIG. 2. — Illustration of the principle of the experiment. $P_e^{(1)}$ is *a*) measured, then *b*) calibrated, by comparison with a known polarization $P_e^{(2)}$.

of the incident beam is changed from linear to circular ; 2) the electric field modulation frequency is divided by two, so that the signal one is now looking for, namely $\mathbf{P}_e^{(2)} . \hat{\mathbf{k}}_f I_f$, varies in time as

$$\cos^2 \frac{\omega_E}{2} t \cos \omega_P t ,$$

and again produces a cross-modulated component, $\cos \omega_E t \cos \omega_P t$, in the photocurrent ; 3) application of a magnetic field $\mathbf{H}$ parallel to $\mathbf{E}_0$ becomes necessary since $\mathbf{P}_e^{(2)}$ is created along $\hat{\mathbf{k}}_i$ and observed in a direction $\hat{\mathbf{k}}_F$ perpendicular to $\hat{\mathbf{k}}_i$. Note that the H-dependence of the signal now exhibits a dispersion shape, instead of an absorption shape in the case of $P_e^{(1)}$. Once the above modifications are done, a measurement of $P_e^{(2)}$ is performed according to a procedure entirely similar to that of $P_e^{(1)}$, except that between two successive sequences of integration, the field is switched from H to $-H$ and the corresponding results are substracted. We obtain :

$$\frac{\mathcal{S}^{(2)}(H) - \mathcal{S}^{(2)}(-H)}{\mathcal{S}_t} = \kappa P_e^{(2)} \frac{2(H/\Delta H)}{1 + (H/\Delta H)^2} . \qquad (12)$$

Several measurements are done for different values of H, and fitted with the variation predicted by eq. (12). This yields the characteristic width ΔH of the Hanle effect and the calibration factor κ. Using eq. (3), (9), (10) and (12), we can now evaluate M_1/E_1 from our experiment :

$$\left| \frac{M_1}{E_1} \right| \simeq \left| \frac{P_e^{(1)}}{P_e^{(2)}} \right| \cdot \left| \frac{\beta}{\alpha} \right| ,$$

with

$$\left| \frac{P_e^{(1)}}{P_e^{(2)}} \right| = \left| \frac{\mathcal{S}^{(1)}}{\mathcal{S}^{(2)}(\Delta H) - \mathcal{S}^{(2)}(-\Delta H)} \right| .$$

In an electric field of 1 000 V/cm, we obtain :

$$\left| \frac{P_e^{(1)}}{P_e^{(2)}} \right| = (2.32 \pm 0.13)\, 10^{-2} . \qquad (13)$$

After combination with the experimental results (8) and (11) this yields :

$$M_1/E_1 = -(2.62 \pm 0.21)\, 10^{-3} \text{ in a field } E_0 = 1\,000 \text{ V/cm} . \qquad (14)$$

The magnetic dipole amplitude is thus found to be equal to the electric dipole amplitude induced by an external field of 2.62 ± 0.21 V/cm. Using the theoretical value of E_1 reported in eq. (5) and the same phase convention, we obtain the transition magnetic moment :

$$cM_1 = \langle 7S, m_S = \tfrac{1}{2} | \mu_z | 6S, m_S = \tfrac{1}{2} \rangle = -(4.24 \pm 0.34)\, 10^{-5} | \mu_B | . \qquad (15)$$

This result can also be expressed in terms of the oscillator strength : $f_{6S-7S} = 4.05 \times 10^{-15}$, and of the single-photon M_1 emission rate

$$\Gamma_{7S-6S} = 0.93 \times 10^{-6} \text{ s}^{-1} .$$

The error quoted in the preceding results (eq. (13) to (15)) represents twice the standard deviation of

our measurements. For cM_1 we have *not* included the error on the theoretical value of E_1 ($< 15\,\%$). We think we have avoided systematic errors connected with phase adjustment of the lock-in amplifiers, with the calibration of the electric field, with possible imperfections in the relative orientation of $\hat{\mathbf{k}}_i$, $\hat{\mathbf{k}}_f$, $\mathbf{E}_0$ and $\mathbf{H}$, and with the imperfection of the incident circular polarization for the measurement of $P_e^{(2)}$. Also, as our determination involves only ratios of similar quantities, the incertainty in the exact vapour density and on the depolarization coefficient κ should constitute no problem. However, it is worth noting that if the relaxation in the upper state were anisotropic in a plane perpendicular to $\mathbf{H}$ as a result, for instance, of the velocity dependence of the laser excitation process, then κ would become different in eq. (10) and (12), and our measurement would not completely eliminate the depolarization effect. However, the source of collision-induced depolarization relevant to our problem is expected to be the collisions between 7S and 6S atoms involving an exchange of excitation, i.e. an exchange of radial quantum numbers, and there is no obvious reason to suppose that the depolarization cross-section might depend on the orientation of the initial spin with respect to the relative velocity of the two atoms; so we do not believe our measurement to be affected in such a way. This is actually confirmed by the results of measurements performed at two vapour densities differing by a factor 4.

In conclusion, magnetic electric dipole interference, by giving rise to the upper state polarization, is a very sensitive tool for measuring quite a weak transition amplitude that might otherwise be obscured in a broad background; it should prove to be useful for several other forbidden transitions. Furthermore, the comparison of this atomic polarization for the two opposite states of circular polarization of the exciting beam provides a rather sensitive way of testing possible parity violation, induced in atomic transitions by weak neutral currents [5], this is presently being followed up [7].

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