Level-crossing spectroscopy of the 7, 9, and 10D states of Cs in an external electric field

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ABSTRACT

We discuss experimental and theoretical studies of coherent excitation of magnetic sublevels in *n*D states of cesium that cross in an external electric field. Crossings of m_F magnetic sublevels of hyperfine F levels with $\Delta m_F = \pm 2$ lead to resonances in the linearly polarized laser induced fluorescence, while crossings with $\Delta m_F = \pm 1$ lead to resonances in the circularly polarized laser induced fluorescence. These resonances can be exploited to observe alignment to orientation conversion. From the level crossing signals it is possible to measure atomic properties, such as the tensor polarizability α_2 and the hyperfine constant A. Alignment to orientation conversion involves the deformation of the spatial distribution of an atom's angular momentum.

Keywords: Level-crossing spectroscopy, alignment to orientation conversion, cesium, two-step excitation, polarizability

1. INTRODUCTION

Level-crossing spectroscopy is a simple, yet effective method for investigating the structure of atomic levels. Electric-field-induced level-crossing spectroscopy has not been exploited as extensively as magnetic-field-induced level-crossing spectroscopy to determine atomic properties, because a homogenous electric field is harder to produce and measure than a magnetic field that causes a comparable magnetic sublevel splitting. The first experimental observation of electric-field-induced magnetic sublevel (m_F) crossing within a hyperfine manifold at non-zero electric field was reported in 1966 by Khadjavi et al.[1] Later, this method was applied to measure the tensor polarizabilities α_2 of the excited states of alkali atoms.[2,3]

Coherently excited magnetic sublevels emit light with a characteristic spatial intensity and polarization distribution. As an external electric field is applied and scanned, coherences that exist at zero electric field are destroyed, and can be partially restored at points where hyperfine sublevels cross (see Fig. 1). As coherences are created and destroyed, the angular distribution of fluorescence of definite polarization changes. Measuring these changes sheds light on the structure of the hyperfine sublevels.

Level crossings of different Δm_F must be studied in slightly different ways. Level-crossing points with $\Delta m_F = \pm 2$ are used in the classic level-crossing experiments, in which linearly polarized light excites the atoms and linearly polarized fluorescence is observed. The linearly polarized light, viewed as a superposition of right- and left-circularly polarized light, can coherently excite levels with $\Delta m_F = \pm 2$. The creation and destruction of these coherences leads to resonance signals as the electric field is scanned.

Level crossings with $\Delta m_F = \pm 1$ make it possible to observe an interesting effect called alignment to orientation conversion in an electric field. Atoms are excited with linearly polarized light, which normally produces an aligned ensemble of atoms, i.e. an ensemble in which the atom has a net electric quadruple moment but does not have a net magnetic dipole moment. However, the presence of an external field at an angle to the polarization of the exciting radiation that is neither zero nor $\pi/2$ allows coherences with $\Delta m_F = \pm 1$ to be created. These coherences created by the light are still components of the alignment created at the angle to the external electric field. The result of the evolution of these coherences in the external electric field is the creation of the orientation of the angular momentum, i.e. an angular momentum distribution with a net magnetic dipole moment. Such alignment to orientation conversion is manifested by the observation of circularly polarized light. This orientation is created in a direction that is perpendicular to the plane which contains light polarization vector **E** and external electric field vector $\boldsymbol{\mathcal{E}}$.

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2. OVERVIEW OF EXPERIMENTS

In our experiments we studied cesium vapor contained in a glass cell at room temperature. Electric fields up to 2400 V/cm were applied via Stark electrodes located inside the cell. In our experiments we used two different cesium cells whose electrodes were separated by 2.5 mm and 5 mm. The experiments are described in detail in.[4,5] In order to populate n(D) states, we applied two-step laser excitation (see Fig. 2). For the first step, to excite the transition $6S_{1/2} \rightarrow 6P_{3/2}$, we used a diode laser (LD-0850-100sm laser diode). In the second step we used either a diode laser (Hitachi HL6738MG laser diode) to excite the $7D_{3/2,5/2}$ states or a Coherent CR699-21 ring dye laser with Rhodamine 6G dye to excite the 9 and $10D_{3/2,5/2}$ states. We observed the laser induced fluorescence (LIF) from the $nD_{3/2}$ states to the the $6P_{1/2}$ state (see Fig. 2a) and from the $nD_{5/2}$ states back to the $6P_{3/2}$ state (see Fig. 2b). When the $nD_{3/2}$ states were studied, the atoms were excited from the $F = 4$ hyperfine structure (hfs) component of the ground state. In the case of the $nD_{5/2}$ states, the atoms were excited from the $F = 3$ hfs component of the ground state. Different initial F states were used in order to populate those hfs components of the final nD state that contain level crossings and to avoid the excitation of the hfs components which contain no level crossings and thus contribute only background.

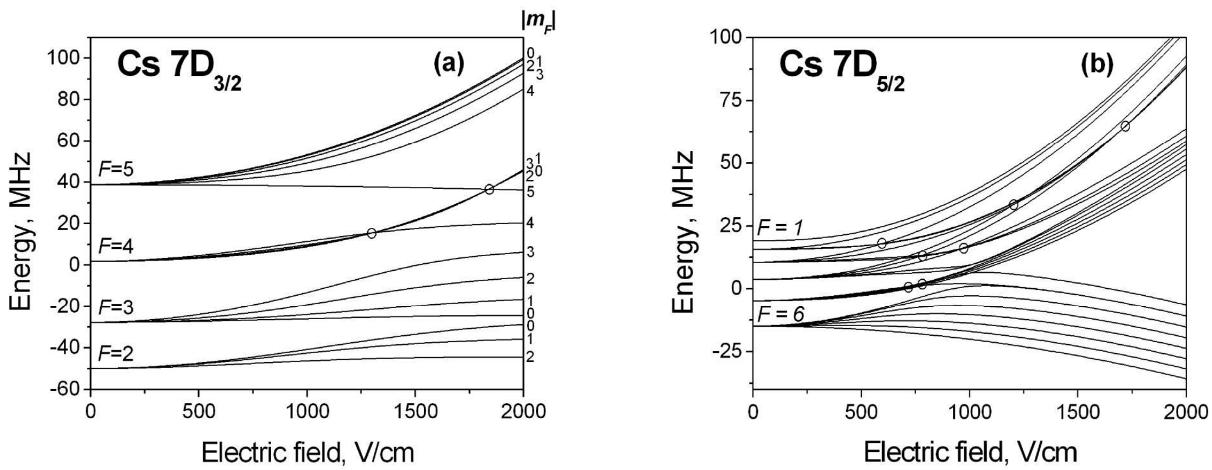

Fig. 1. Hyperfine level splitting diagram in external electric field for the (a) $7D_{3/2}$ and (b) $7D_{5/2}$ states of Cs. Circles indicate level crossings with $\Delta m_F = \pm 2$.

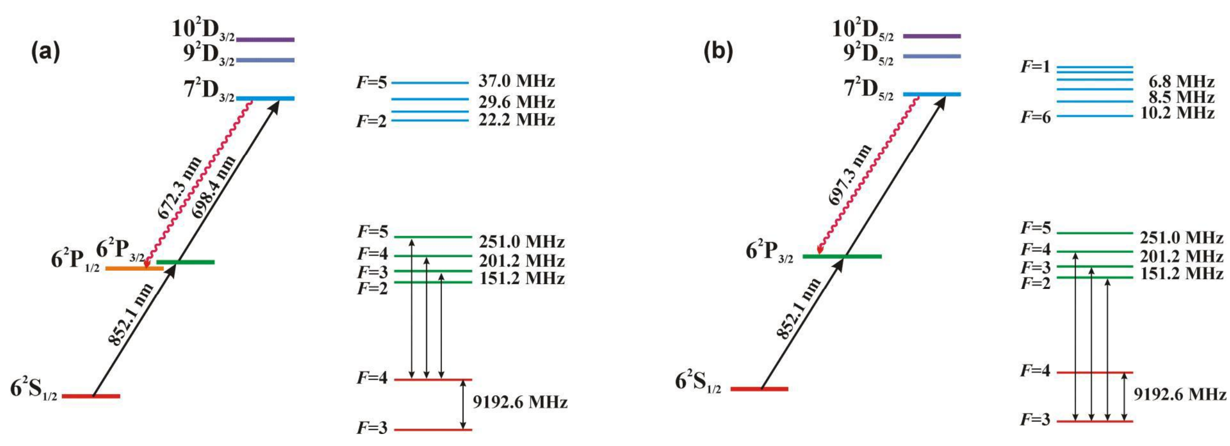

Fig. 2. Cesium energy levels and excitation-observation schemes for the (a) $nD_{3/2}$ states and (b) $nD_{5/2}$ states

To create coherence in excited atomic states, the relative orientation of the electric field $\boldsymbol{\mathcal{E}}$ and the polarization of the exciting lasers $\mathbf{E_1}$ and $\mathbf{E_2}$ is of great importance. In order to observe level-crossing (LC) resonance signals, which were caused by coherence between magnetic sublevels with $\Delta m_F = \pm 2$, the relative orientation of the electric field and the polarization vectors of the lasers was the following: the first laser, with polarization vector $\mathbf{E_1}$, was linearly polarized parallel to the external dc electric field $\boldsymbol{\mathcal{E}}$ ($\boldsymbol{\mathcal{E}} \| \mathbf{z}$); the second laser, with polarization vector $\mathbf{E_2}$, was sent in a counter-propagating direction and polarized perpendicular to the electric field $\boldsymbol{\mathcal{E}}$ ($\mathbf{E_2} \| \mathbf{y}$, see Fig. 3a). The LIF was observed along the electric field direction, and the dependence on the electric field strength of the LIF components I_x and I_y were studied.

In order to observe alignment to orientation conversion (AOC), which is caused by coherence between magnetic sublevels with $\Delta m_F = \pm 1$, the relative orientation of the electric field and the lasers' polarization vectors was the following: the first laser, with polarization vector $\mathbf{E_1}$, was linearly polarized parallel to the external dc electric field $\boldsymbol{\mathcal{E}}$ ($\boldsymbol{\mathcal{E}} \| \mathbf{z}$); the second laser , with polarization vector $\mathbf{E_2}$, was sent in a counter-propagating direction and polarized at an angle $\pi/4$ with respect to the electric field $\boldsymbol{\mathcal{E}}$ (see Fig. 3b). The LIF was observed along the laser beam propagation direction with help of a pierced mirror. The degree of LIF circularity as a function of the external dc electric field was studied.

Fig. 3. Schematic diagram of the level crossing experiment (a) $\Delta m_F = \pm 2$ and (b) $\Delta m_F = \pm 1$.

Typical measured LC signals are shown for the $7D_{3/2}$, $9D_{3/2}$, and $10D_{5/2}$ states in Figs. 4-6, where the LIF relative intensity is plotted as a function of the electric field strength. The measured signals are represented by dots with error bars and simulations by a solid line. The plotted simulations are based on a correlation analysis of the optical Bloch equations in the case when an atom simultaneously interacts with two laser fields in the presence of an external dc electric field. These simulations are described in detail in.[4] Experimental LC signals are presented for two geometries, which we labeled as *zyx* and *zyy*, where the first and the second letters, *z* and *y,* denote the orientations of the polarization vectors of the first and second lasers, $\mathbf{E_1} \| z$ and $\mathbf{E_2} \| y$, and the third letter, *x* or *y*, denotes the polarization direction of the observed LIF.

The LC signals for the $n\mathrm{D}_{3/2}$ states contain two well pronounced resonances (see Fig. 4, 5). The positions of these resonances correspond to the electric field values at which magnetic sublevels with $\Delta m_F = \pm 2$ cross (see Fig. 1a). The level-crossing positions depend on the tensor polarizability α_2 and on the hfs constants *A* and *B* of the atomic state. Thus, by measuring the resonance position, it is possible to determine either the tensor polarizability or the hfs constant, assuming that one of them is known. For the $n\mathrm{D}_{3/2}$ states of Cs the values of the magnetic dipole hfs constants *A* are known with good precision, and the values of the electric quadrupole hfs constant *B* have been reported to be negligibly small.[6] This state of affairs allowed us to measure the tensor polarizability α_2 of the $7D_{3/2}$ and $9D_{3/2}$ states. The results of α_2 measurements for $n\mathrm{D}_{3/2}$ states are given in Table 1.

The experimental signals for $n\mathrm{D}_{5/2}$ states do not contain well defined resonances as in case of the $n\mathrm{D}_{3/2}$ states (see Fig. 6). The reason is that the $n\mathrm{D}_{5/2}$ state's hyperfine manifold has many closely spaced $\Delta m_F = \pm 2$ magnetic sublevel crossings (see Fig. 1b), and resonance signals from these level crossings overlap. In this case, in order to obtain reliable values of atomic constants, a description of the measured fluorescence dependence on the electric field by a detailed theory is essential. The atomic constants α_2 and *A* can be derived as variable parameters, which fit the experimental signal, of the simulation based on this theory. Such a theory was developed and tested for the $n\mathrm{D}_{3/2}$ states of Cs.[4]

Fig. 7 demonstrates the alignment to orientation conversion signal induced by an external dc electric field for the $9D_{3/2}$ state of Cs. This signal can be interpreted also as the crossing of magnetic sublevels with $\Delta m_F = \pm 1$ within the hyperfine manifold and reflects the coherent evolution of these sublevels under the influence of the external electric field.[5] The solid line represents the results of calculations based on a theoretical model developed to solve the optical Bloch equations.[4] The insets in Fig. 7 help to visualize the angular momentum probability distribution at zero electric field and at the electric field value for which the degree of orientation was at a maximum. It is apparent from the figure that the theory is able to describe the experimental signals quite well. Such deformations of atomic angular momentum distributions could be a background in some sensitive searches for a permanent electric dipole moment of the electron, and hence the possibility to calculate precisely the deformations for various experimental conditions could be useful.[5]

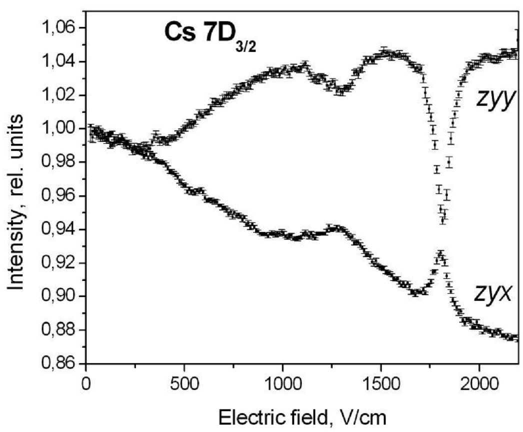

Fig. 4. Experimentally observed level crossing signals ($\Delta m_F = \pm 2$) for the $7D_{3/2}$ state of Cs, *zyy* and *zyx* geometries.

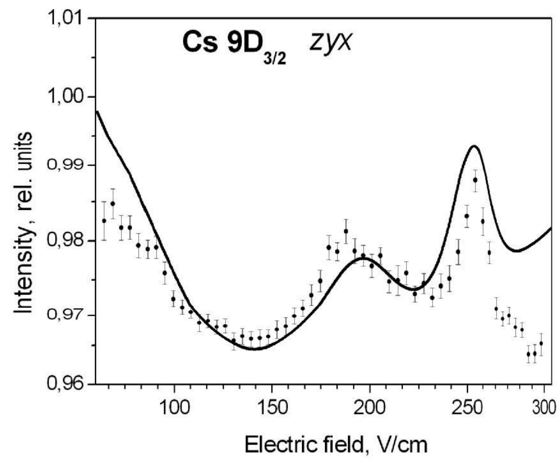

Fig. 5. Level crossing signal ($\Delta m_F = \pm 2$) for the $9D_{3/2}$ state of Cs, *zyx* geometry. Dots: measurements, solid line: simulation.

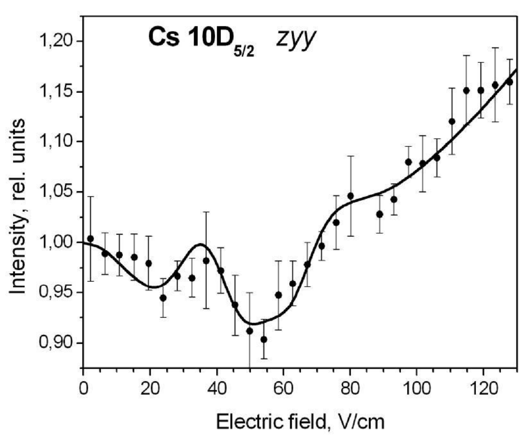

Fig. 6. Level crossing signals ($\Delta m_F = \pm 2$) for the $10D_{5/2}$ state of Cs, *zyy* geometry. Dots: measurements, solid line: simulation.

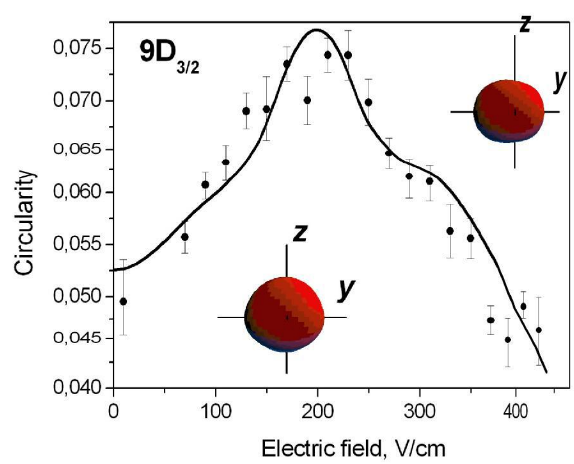

Fig. 7. Alignment to orientation conversion signal ($\Delta m_F = \pm 1$) for the $9D_{3/2}$ state of Cs. Dots: measurements, solid line: simulation. Insets depict the atomic angular momentum distribution.

Table. 1 Tensor polarizability values for 7 and $9D_{3/2}$ states obtained from the level-crossing experiment in an external electric field.[4]

Cesium atomic state	Hyperfine constant A, (MHz)	Tensor polarizability α_2, (a_0^3)
$7D_{3/2}$	7.4(2) [6]	$7.45(20) \times 10^4$
$9D_{3/2}$	2.35(4) [6]	$1.183(35) \times 10^6$

3. CONCLUSIONS

Level-crossing spectroscopy within the hyperfine manifold in an external dc electric field can be used as an experimental method to determine atomic properties such as the tensor polarizability α_2 or the hyperfine structure constant A. For the $n\mathrm{D}_{3/2}$ states, values for one of these constants can be obtained from the positions of the level-crossing resonances if the other constant is known. The $n\mathrm{D}_{5/2}$ states in Cs, however, contain so many level crossings that well defined resonances can no longer be observed. In this case, detailed signal simulations are essential for extracting the atomic constants by fitting experimental signals with the results of simulations.

Accurate theoretical descriptions of signals for different types of level-crossing experiments can be obtained. In order to improve further the agreement with experiment, work is in progress to include averaging over the Doppler profile in the simulations. This improvement should make it possible to obtain atomic properties with even greater precision and confidence.

The electric-field dependence of the signals from the experiments described above suggests the possibility of using the signals to map electric field distributions by optical methods. Work is currently in progress to evaluate the effectiveness of this approach to mapping electric field distribution

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