Electric Field Induced Symmetry Breaking of Angular Momentum Distribution in Atoms

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We report the experimental observation of alignment to orientation conversion in the 7D_{3/2} and 9D_{3/2} states of Cs in the presence of an external dc electric field, and without the influence of magnetic fields or atomic collisions. Initial alignment of angular momentum states was created by two-step excitation with linearly polarized laser radiation. The appearance of transverse orientation of angular momentum was confirmed by the observation of circularly polarized light. We present experimentally measured signals and compare them with the results of a detailed theoretical model based on the optical Bloch equations.

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Atomic physics experiments are able to place sensitive limits on the permanent electric dipole moment (EDM) of the electron because highly polarizable atoms can generate internal electric fields that are orders of magnitude higher than can be achieved for free particles. Such limits on the electron EDM are of great interest to fundamental physics because a permanent EDM of a fundamental particle implies CP violation as long as the CPT theorem is assumed to hold. Such a CP violation would be a signature of new physics beyond the standard model. The most sensitive upper limit to date on the electron EDM has been achieved by searching for a small precession around an external electric field. In this case, collisions with electrons served as a source of initial alignment, which was then transformed into orientation by a combination of static electric and magnetic fields.

Angular momentum alignment means that the population of atomic sublevels $m_J$ varies with $|m_J|$, but $+m_J$ and $-m_J$ levels are populated equally. When the $+m_J$ and $-m_J$ levels are unequally populated, the ensemble is oriented. One can describe the anisotropic spatial distribution of angular momenta $J$ in an ensemble by an atomic density matrix $\rho$. An ensemble is described as aligned if the distribution of angular momenta contains a net electric quadrupole moment or oriented if there is a net magnetic dipole moment.

The very first theoretical works suggested that AOC may be induced in atoms by a magnetic field gradient or by anisotropic collisions in which the angle between the collision axis and the alignment axis differs from 0 and $\pi/2$. Experimental studies of AOC in atoms in a purely electric field without any need for magnetic fields or collisions only were used to study AOC in Rb, Na, and Cs. When a strong pumping laser field excites atoms in the presence of a magnetic field, the interaction of the dynamic Stark effect with the magnetic field was shown to lead to AOC, first in Hg and then in other elements. In particular, a sizeable degree of circular polarization, caused by AOC, was observed in the $5d6p^1P$ state of Ba in a femtosecond regime due to the combined effects of a static magnetic field and an electric field from a pulsed laser. Lombardi et al. measured the appearance of orientation in the $4^1D_2$ state of helium using a capacitative electrodeless helium discharge. In this case, collisions with electrons served as a source of initial alignment, which was then transformed into orientation by a combination of static electric and magnetic fields. Similar experiments were performed by Elbel et al. Since then, there have been experiments that have shown how AOC can be caused in atomic systems by magnetic field gradients, collisions, and combined electric and magnetic fields. However, no experimental studies of AOC in atoms in a purely electric field have been reported.

It has been predicted that the quadratic Stark effect in the presence of an electric field only could cause AOC without any need for magnetic fields or collisions. In general, linearly polarized light is expected to
produce alignment, but not orientation. However, initial alignment produced by linearly polarized light can be converted into transverse orientation when the aligned atoms are placed in a perturbing electric field that makes an angle with the initial alignment axis different from 0 or $\pi/2$ and the quadratic Stark effect causes levels with $\Delta m = \pm 1$ to cross.

In this letter we confirm these predictions with quantitative measurements, which we compare with the results of a simulation.

In our experiment we studied AOC in the $7D_{3/2}$ and $9D_{3/2}$ states of Cs. Figure 1 shows the hyperfine level splitting in an external electric field for these states, which are calculated by diagonalizing in an uncoupled basis the Hamiltonian, which includes the hyperfine and Stark terms [24]. To compute this Hamiltonian, we use the following atomic constants: hyperfine constant $A = 7.4(2)$ MHz [25], scalar polarizability $\alpha_0 = -6.0(8) \times 10^4$ [26] in $a_0^3$ units for the $7D_{3/2}$ state, and $A = 2.35(4)$ MHz [25], $\alpha_0 = -1.45(12) \times 10^6$ [28], and $\alpha_2 = 1.183(35) \times 10^6$ [27] for the $9D_{3/2}$ state.

The circle in each diagram marks a crossing of magnetic sublevels with $\Delta m = \pm 1$. Two sublevels with $\Delta m = \pm 1$ can be simultaneously excited with linearly polarized light when they are degenerate in energy and there is a non-zero angle between the polarization vector of the exciting radiation and the quantization axis, i.e., the electric field axis. The relative orientations of the electric field $\mathbf{E}$ and the laser polarization vectors $\mathbf{E}_1$ and $\mathbf{E}_2$ and the laser induced fluorescence (LIF) observation direction are shown in Fig. 2. In this setup, the appearance of transverse orientation, that is, orientation perpendicular to the quantization axis, can be detected by observing circularly polarized light in a direction perpendicular to $\mathbf{E}$ as well as $\mathbf{E}_1$ and $\mathbf{E}_2$.

We make measurements on cesium vapor contained in a glass cell at room temperature. In order to reach the $7D_{3/2}$ and $9D_{3/2}$ states of cesium we use two-step laser excitation with counterpropagating laser beams (see Fig. 3). In the first step atoms are excited to the $6P_{3/2}$ state by a diode laser (LD-0850-100sm laser diode), which is linearly polarized along the external dc electric field $\mathbf{E}$ direction ($\mathbf{E} \parallel \mathbf{E}_1$). In the second step, we use either a diode laser (Hitachi HL6738MG laser diode) to induce the $6P_{3/2} \rightarrow 7D_{3/2}$ transition, or a Coherent CR699-21 ring dye laser with Rodamin 6G dye to induce the $6P_{3/2} \rightarrow 9D_{3/2}$ transition. The polarization vector of the second laser $\mathbf{E}_2$ makes an angle of $\pi/4$ with respect to that of the first $\mathbf{E}_1$ and the electric field $\mathbf{E}$ (see Fig. 2).

An electric field is applied via polished stainless steel electrodes of 25 mm diameter located inside the cell and separated by a 5 mm gap. The LIF at the $nD_{3/2} \rightarrow 6P_{1/2}$ transition is observed collinearly to the
laser beams with the help of a pierced mirror. Before entering a monochromator, the LIF passes through a $\lambda/4$ plate, which converts circularly polarized light into linearly polarized light, allowing us to measure the degree of circularity $C$ defined as

$$C = \frac{I(E_{\text{right}}) - I(E_{\text{left}})}{I(E_{\text{right}}) + I(E_{\text{left}})},$$

where $I(E_{\text{right}})$ and $I(E_{\text{left}})$ are the intensities of the right and left circularly polarized LIF.

The signal is detected by a photomultiplier tube in photon counting mode and recorded on a PC together with the electrode voltage. The voltage is applied in discrete steps. During each step, the number of photons is counted for $E_{\text{right}}$ and $E_{\text{left}}$.

The results of our measurements are shown as dots in Fig. 4. The circularity $C$ reaches a maximum near the $\Delta m = \pm 1$ crossing (circled in Fig. 1). A value of $C$ as high as 10% is observed in the case of the 7D$_{3/2}$ state. A small orientation appears at zero electric field because the linewidth of the second laser is sufficiently broad to excite coherently magnetic sublevels belonging to different $F$ states. This effect is more pronounced in the 9D$_{3/2}$ state because of the smaller hyperfine level splittings. Solid lines show the result of simulations. Our theoretical treatment builds on earlier models developed to solve the rate equations for Zeeman coherences for single-step [29] and double-step [27] laser excitation of atoms.

The model assumes that the atoms move classically and are excited by the laser radiation at the internal transitions. In this case, the internal dynamics of the atom can be described by a semiclassical atomic density matrix $\rho$ parametrized by the classical coordinates of the atomic center of mass. To obtain the density matrix, we must solve the optical Bloch equations (see, for example, Ref. [30])

$$i\hbar \frac{\partial \rho}{\partial t} = [\hat{H}, \rho] + i\hbar \hat{R}\rho.$$  

The Hamiltonian $\hat{H}$ includes the unperturbed Hamiltonian for the hyperfine interaction, the dipole interaction operator, and the dc Stark operator. The operator $\hat{R}$ describes relaxation and includes both spontaneous emission and transit relaxation. We assume that the density of atoms is sufficiently low so that different velocity groups of thermally moving atoms do not interact. The model also accounts for nonlinear effects such as the ac Stark effect even though they are small in our experiment because the laser power was low.

We apply the rotating wave approximation for multilevel systems [31] to eliminate oscillations with optical frequencies. Since the time scale of the experiment is large compared to the characteristic phase fluctuation time of the lasers, we take a statistical average over the fluctuating phases [29]. We assume that both lasers are uncorrelated, so that we can apply the “decorrelation approximation” [29, 32] to obtain a system of equations, which, when solved, yields the density matrix of the $nD_{3/2}$ state, from which the fluorescence intensities in each polarization are obtained.

We obtained the values of those simulation parameters that could not be controlled precisely in the experiment by fitting simulations and measurements of level crossing signals of the same (7,9)D$_{3/2}$ Cs states, which were measured under the same experimental conditions [27]. These parameters included the spectral widths of the laser radiation and the detuning with respect to the exact hyperfine transition frequencies.

The excellent agreement between experiment and theory (see Fig. 4) demonstrates the validity of our theoretical approach.

To convey an intuitive understanding of the AOC effect, Fig. 4 contain inserts which help to visualize the angular momentum distributions at zero electric field, and at the electric field value for which the degree of orientation was at a maximum. The angular momentum distribution is visualized as a surface whose distance from the origin in proportional to the probability that the angu-
lar momentum of an atom in the ensemble points toward that point on the surface. This probability is computed from the components of the density matrix $\rho_{mm'}$. The theoretical model involves computing the density matrix $\rho^{FF'}_{MM'}$ for the manifold of hyperfine levels. This matrix describes the population of certain $F$ states, as well as the Zeeman coherences inside those states and between different $F$ states. Since we do not resolve spectrally the $nD$ state hyperfine components that are separated by $\sim 10$ MHz, it is convenient in the final analysis to pass to the overall density matrix for a fine structure level. To this end, we need to contract the fine structure density matrix over the nuclear spin $I$ projections $\mu, \mu'$ and sum over the hyperfine structure components with the expansion coefficients of the fine structure wave functions over the hyperfine structure wave functions

$$\rho_{mm'} = \sum_{\mu} \rho_{mm\mu\mu'} \delta_{\mu\mu'} = \sum_{FF'MM'} \rho_{MM'}^{FF'} \sum_{\mu} C_{FM1\mu}^{F'} C_{M1\mu}^{F}.$$

(3)

In the absence of the external field these expansion coefficients are the Clebsch-Gordan coefficients $C_{FM1\mu}^{F'}$. If the electric field is present, the Clebsch-Gordan coefficients must be replaced by the expansion coefficients that can be obtained by diagonalizing the Stark Hamiltonian.

In summary, we have observed AOC produced by the quadratic Stark effect in an external electric field without the need of any magnetic fields or collisions. With the light $E$ vectors and the external dc electric field $E$ forming an angle of $\pi/4$, transverse orientation perpendicular to the $E-E$-plane appeared, and gave rise to LIF signals with a degree of circularity up to 10%. In order to search for the electron EDM in atomic or molecular systems, it is often crucial to ensure that the dc electric field, the light polarization vectors, and the residual magnetic field are exactly parallel or orthogonal to each other. As experiments push the limit even lower, this requirement will be hard to achieve with the necessary accuracy, and so AOC could introduce a possible background. Our investigation shows that AOC can be understood very well and corrections could be made, if necessary.

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