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. The effect is odd under time reversal and should be taken into account in ever more sensitive searches for an electron electric dipole moment.

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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 [1,2]. 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 [3]. 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 of the angular momentum distribution of an ensemble of atoms [4]. Various sophisticated experimental techniques prevent angular momentum precession caused by mechanisms other than an EDM from contaminating the signal. One such phenomenon, known as alignment to orientation conversion (AOC), is known to deform the atomic and molecular angular momentum distributions under the influence of combined electric and magnetic fields or collisions. Moreover, it has been shown theoretically [5,6] that AOC can be induced by a purely electric field without the need for magnetic fields or collisions whenever the initial alignment is not exactly perpendicular or parallel to the external electric field. In this Letter, we report the experimental observation of AOC in the presence of only an electric field. Our measured signals are described accurately by a detailed theoretical model, which could be of interest in studying potential backgrounds for electron EDM searches in atomic or molecular systems.

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 $\mathbf{J}$ in an ensemble by an atomic density matrix ρ [7]. 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. Orientation can be classified as longitudinal or transverse with reference to the symmetry axis.

An atom placed in an electric field $\boldsymbol{\mathcal{E}}$ will become polarized, and, in general, the polarization will have a component parallel to the angular momentum $\mathbf{J}$. This component leads to a torque on the angular momentum proportional to $(\boldsymbol{\mathcal{E}} \cdot \mathbf{J})(\boldsymbol{\mathcal{E}} \times \mathbf{J})$ that distorts the atomic angular momentum distribution in the plane perpendicular to the electric field and the axis of initial alignment [8]. Thus, 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 $\pi/2$. The effect can be understood also as coherent evolution of different hyperfine sublevels with $\Delta m = \pm 1$, which can cross under the influence of an external electric field [9].

The breaking of the reflection symmetry of an initially aligned population of atoms by converting the alignment into transverse orientation has been a subject of theoretical and experimental investigation since the 1960s, beginning with the work of [10–13]. The very first theoretical works suggested that AOC may be induced in atoms by a magnetic field gradient [10] or by anisotropic collisions in which the angle between the collision axis and the alignment axis differs from 0 and $\pi/2$ [11,12]. These latter predictions were confirmed by [14,15]. Orthogonal static electric and magnetic fields were used to study AOC in Ba and Cs [16], while only magnetic fields were used to study AOC in Rb [17,18] and Na [19]. When a strong pumping laser field excites atoms in the presence of a magnetic field, the dynamic Stark effect in the presence of the magnetic field was shown to lead to AOC, first in Hg [20] and then in other elements [21,22]. In particular, a sizeable degree of circular polarization (around 15%), 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 [22]. Lombardi *et al.* [13,23] measured the appearance of ori-

entation 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.* [24]. 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.

In our experiment we studied AOC in the $7D_{3/2}$ and $9D_{3/2}$ states of Cs in the presence of a purely electric field. 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 [25]. To compute this Hamiltonian, we use the following atomic constants: hyperfine constant $A = 7.4(2)$ MHz [26], scalar polarizability $\alpha_0 = -6.0(8) \times 10^4$ [27], and tensor polarizability $\alpha_2 = 7.45(20) \times 10^4$ [28] in a_0^3 units for the $7D_{3/2}$ state; and $A = 2.35(4)$ MHz [26], $\alpha_0 = -1.45(12) \times 10^6$ [29], and $\alpha_2 = 1.183(35) \times 10^6$ [28] 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 nonzero 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 $\boldsymbol{\mathcal{E}}$ 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 $\boldsymbol{\mathcal{E}}$ 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 $\boldsymbol{\mathcal{E}}$ direction ($\boldsymbol{\mathcal{E}} \parallel \mathbf{E}_1$). 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 rhodamine 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 $\boldsymbol{\mathcal{E}}$ (see Fig. 2).

An electric field is applied via polished stainless steel Stark 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 followed by a linear polarizer, which converts circularly polarized light into linearly polarized light, allowing us to measure the degree of circularity C defined as

$$C = \frac{I(E_{\text{right}}) - I(E_{\text{left}})}{I(E_{\text{right}}) + I(E_{\text{left}})}, \tag{1}$$

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 personal computer together with the electrode voltage. The voltage is applied in discrete steps. During each step, the number of photons is counted for E_{right} and E_{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

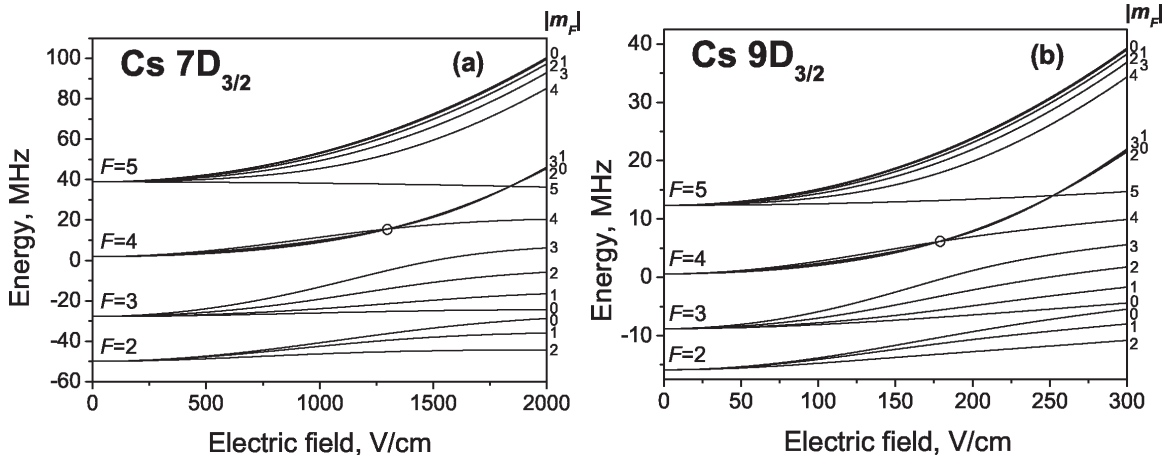

FIG. 1. Hyperfine level splitting diagram in an external electric field for the (a) $7D_{3/2}$ and (b) $9D_{3/2}$ states of Cs.

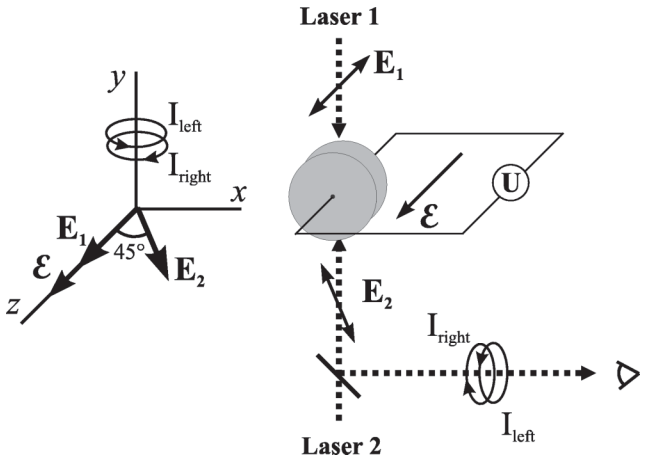

FIG. 2. Experimental geometry.

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 [30] and double-step [28] 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 ρ 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. [31])

$$i\hbar\frac{\partial\rho}{\partial t} = [\hat{H}, \rho] + i\hbar\hat{R}\rho. \tag{2}$$

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

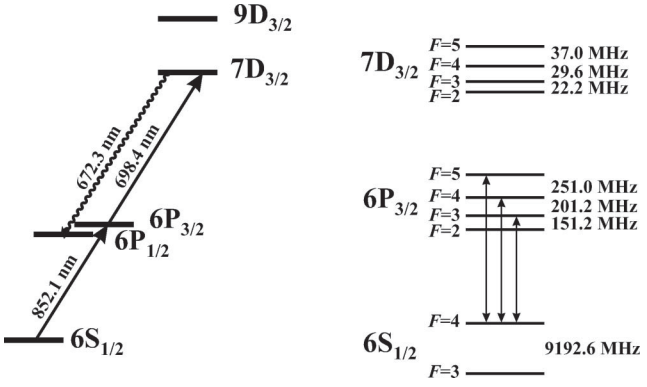

FIG. 3. Cs energy levels and excitation-observation scheme.

though they are small in our experiment because the laser power is low.

We apply the rotating wave approximation for multilevel systems [32] 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 [30]. We assume that both lasers are uncorrelated, so that we can apply the "decorrelation approximation" [30,33] 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 [28]. 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 contains insets 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 is proportional to the probability that the angular 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 $\rho_{mm'}$ [9,34]. 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 μ, μ' and sum over the hyperfine structure components with the expansion coefficients of the fine structure wave functions over the hyperfine structure wave functions

$$\rho_{mm'} = \sum_{\mu}\rho_{m\mu m'\mu'}\delta_{\mu\mu'} = \sum_{FF'MM'}\rho^{FF'}_{MM'}\sum_{\mu}C^{FM}_{JmI\mu}C^{F'M'}_{Jm'I\mu}. \tag{3}$$

In the absence of the external field these expansion coefficients are the Clebsch-Gordan coefficients $C^{FM}_{JmI\mu}$. 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.

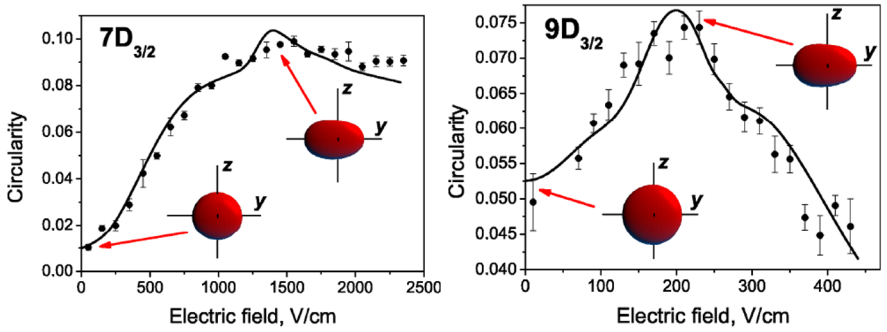

FIG. 4 (color online). Degree of LIF circularity C as a function of electric field for the $7D_{3/2}$ and $9D_{3/2}$ states of Cs. Points: experiment. Solid line: simulation. Insets depict the atomic angular momentum distribution.

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 $\boldsymbol{\mathcal{E}}$ forming an angle of $\pi/4$, transverse orientation perpendicular to the $\mathbf{E}\boldsymbol{\mathcal{E}}$ plane appeared and gave rise to LIF signals with a degree of circularity up to 10%. The AOC signal is of order $\alpha(\boldsymbol{\mathcal{E}} \cdot \mathbf{J})(\boldsymbol{\mathcal{E}} \times \mathbf{J})$, where α is the polarizability. The created angular momentum orientation is odd under time reversal, and although it is even under parity inversion, it nevertheless should be taken into account as searches for an electron EDM become ever more sensitive. AOC could substantially alter the nonlinear optical rotation in the presence of an electric field that is used as a signature for an EDM [21]. AOC could mimic an EDM, if, for example, the electric fields were not perfectly reversed [35]. The magnitude of the deformation of the angular momentum caused by AOC is proportional to $\sin(2\theta)$, where θ is the angle between the electric field and the alignment axis [9]. Our investigation shows that AOC can be understood very well and corrections could be made, if necessary.

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