

# Rotation properties of multipole moments in atomic sublevel spectroscopy

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The spatial orientation of atomic multipole moments can be observed optically with resonant polarized light. We demonstrate the procedure in the ground state of atomic sodium, using optical pumping to polarize the angular momentum substates. We show how the resulting information can be used to distinguish among signal contributions originating from different multipole moments.

Optical pumping of a resonant atomic medium is an efficient method to create atomic multipole moments. If the cylindrical symmetry of the optical pumping experiment is broken by a transverse magnetic field, atomic multipole moments of all possible orders can be excited. Some of these multipole moments modify the optical properties of the medium and can therefore be monitored optically.<sup>1</sup> Direct optical observation of atomic multipole moments is possible and has been reported for dipole moments<sup>1</sup> and quadrupole moments.<sup>2-4</sup> Higher moments are not accessible in direct optical experiments,<sup>2</sup> but indirect evidence has been obtained for moments up to hexadecapoles.<sup>5,6</sup> The methods used for such observations are either zero-field level-crossing experiments<sup>5,6</sup> or Raman-type experiments in a static magnetic field, where the multipole moments can be identified through their precession frequency.<sup>1,2,7</sup> These methods not only facilitate the assignment of the signal contributions to the various multipole moments but also allow us to separate the information in the frequency dimension.

Although direct optical monitoring of higher-order multipole moments is not possible, we recently demonstrated a technique that permits indirect observation of all multipole moments.<sup>8</sup> The technique uses a laser-induced transfer of the relevant spectroscopic information from the invisible higher moments to a sublevel transition that can be observed directly. The data that are obtained by this procedure permit the observation of the free evolution of the system. If these time-domain data are Fourier transformed, one obtains a spectrum whose resonance frequencies correspond to transitions between the different angular momentum substates. In the examples considered here, these substates belong to the electronic ground state and have correspondingly long lifetimes. In the presence of a magnetic field the different atomic multipoles evolve at specific frequencies; therefore their signal contributions can be identified by their resonance frequency.

This method permits the observation of multipole moments of any order and thus makes possible experiments that can completely characterize the state of an atomic system by independently measuring the different multipole moments. In systems with

nonvanishing nuclear spin the number of resonances in such a spectrum can become quite large because every hyperfine multiplet contributes  $(2F + 1)^2$  independent variables. Therefore an unambiguous separation of signal contributions from the individual multipole moments becomes a nontrivial task. In many cases this assignment is greatly facilitated if the signal components can be characterized by additional variables.

In this Letter we demonstrate such a possibility of distinguishing signal contributions arising from different atomic multipole moments. Our method uses directly the rotational transformation properties of irreducible tensor operators: signal components depending on multipole moments of order  $m$  are unchanged if the atomic system is rotated by an angle  $2\pi/m$  but can be distinguished for intermediate rotation angles. By comparing signals from atomic states that differ only by a rotation around the quantization axis, one can distinguish contributions from different atomic multipole moments.

The atomic multipole moments that we investigate here are created in the electronic ground state of atomic sodium. For simplicity, we restrict the analysis to the  $F = 2$  multiplet, where multipole moments up to hexadecapole ( $\Delta m_F = 4$ ) are possible. The atomic medium is placed in a magnetic field oriented perpendicular to the laser beams. A pump laser is used to create the multipole moments, and a second, weak, laser beam monitors their evolution. After being transmitted through the medium, this probe laser beam is passed into a polarization-selective detector,<sup>9</sup> which produces a signal proportional to the atomic magnetic dipole moment component in the direction of the laser beam. Higher-order multipole moments are obtained by a procedure discussed in detail elsewhere.<sup>8</sup>

If the multipole moments are excited by optical pumping with a time-independent pump laser, the steady-state atomic polarization during the pulse has a fixed orientation in space that depends on the optical pumping rate and the magnetic field strength.<sup>9</sup> However, if the pump laser is modulated, it is possible to create multipole moments that rotate around the direction of the magnetic field.<sup>10,11</sup> The precession

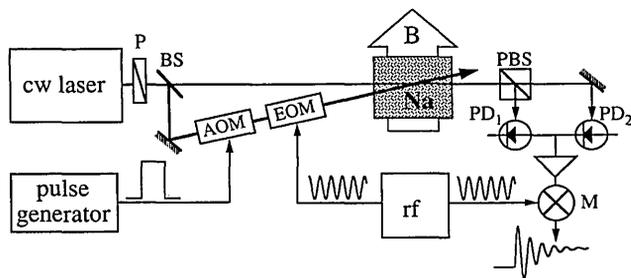


Fig. 1. Schematic of the experimental setup: P, polarizer; BS, beam splitter; AOM, acousto-optic modulator; EOM, electro-optic modulator; rf, radio-frequency synthesizer; B, magnetic field; PBS, polarizing beam splitter; PD's, photodiodes; M, mixer.

frequency during the optical pumping process is equal to the modulation frequency, and the instantaneous orientation of the atomic state can be controlled by the modulation phase<sup>10</sup>: shifting the phase by an angle  $\Delta\phi$  leads directly to a rotation of the resulting atomic state by an angle  $\Delta\phi$ .

This precessing order within the angular momentum substates is conventionally expanded in a basis of multipole moments corresponding to irreducible tensor operators  $T_q^{(k)}$  of rank  $k$  and order  $q$ .<sup>12</sup> In the eigenbase of the angular momentum operator  $F_z$ , these operators involve nonvanishing matrix elements between states whose magnetic quantum numbers differ by  $\Delta m_F = q$ . By definition, tensor operators of order  $q$  are invariant under rotations around the quantization axis by an angle  $2\pi/q$ ; their contributions to the overall signal must therefore also be invariant under such a rotation. More precisely, they depend on the rotation angle  $\phi$  by a phase factor  $\exp(iq\phi)$ . Thus a shift of the phase of the pump laser modulation by  $\Delta\phi$  should change signal contributions from multipole moments of order  $q$  by a factor  $\exp(iq\Delta\phi)$ . This phase dependence provides a signature that permits the identification of signal contributions from different multipole moments.

The experimental setup used for the implementation of this procedure is shown in Fig. 1. The measurements reported here were performed on the  $F = 2$  multiplet of the  $3s^2S_{1/2}$  sodium ground state. The atomic vapor was contained in a 1-cm-long glass cell at a temperature of 200 °C together with 200 mbars of Ar buffer gas. The pressure broadening of 4-GHz full-width at half-height produced a homogeneous optical resonance line. The light source was a single-mode cw ring dye laser with a short-term linewidth of less than 500 kHz. The laser frequency was set near the Na  $D_1$  line ( $\lambda = 589.6$  nm) with a detuning  $\Delta/2\pi = 12$  GHz above the center of the (pressure-shifted) resonance line, optimized for efficient detection of the higher-order multipole moments with  $|q| = |\Delta m_F| > 1$ . The laser beam was split into a pump beam with an intensity of  $\sim 150$  mW/mm<sup>2</sup> and a probe beam with an intensity of  $\sim 0.6$  mW/mm<sup>2</sup>. The two beams overlapped in the sample region at an angle of some 10 mrad. The optical pulses were generated by an acousto-optic modulator driven by

a computer-controlled digital delay generator. With an electro-optic modulator, the polarization of the pump laser beam was modulated between opposite circular polarizations at a frequency of  $\omega_m/2\pi = 5.2$  MHz.<sup>11</sup>

After propagating through the vapor cell, the probe beam passed into a polarization-selective detector,<sup>9</sup> which measured the circular birefringence of the sample. The frequency of this signal was downconverted with a double-balanced mixer with the modulation frequency used as the reference.<sup>10</sup> The signal was then digitized and transferred to a computer for data analysis and plotting. An example of a resulting spectrum is shown in Fig. 2. We obtained the data by measuring the signal as a function of time after the pump laser pulse. We Fourier transformed these time-domain data to separate the signal contributions from different multipole moments in the frequency dimensions, as shown in Fig. 2. The main resonance lines are identified in the figure and assigned to transitions between angular momentum substates of the Na ground state: the numbers identifying the resonance lines indicate the quantum numbers ( $F, m_F \leftrightarrow F', m_{F'}$ ) of the sublevels connected by the individual transitions. The separation of the lines within each multiplet is due to the quadratic Zeeman effect.<sup>13</sup> Therefore this representation of the signal permits a distinction of the contributions from different multipole moments: each resonance corresponds to a tensorial component of order  $q = m_F - m_{F'}$ . Nevertheless, the severe overlap among the various resonance lines makes the distinction difficult and sometimes impossible, especially for a small signal whose resonance frequency is near that of a large resonance line.

We implemented the procedure described above by comparing spectra of this type corresponding to different orientations of the atomic system. For this purpose we performed a sequence of four experiments with different modulation phases during the optical pumping process, but we kept the reference phase for the phase-sensitive detection constant. We expect

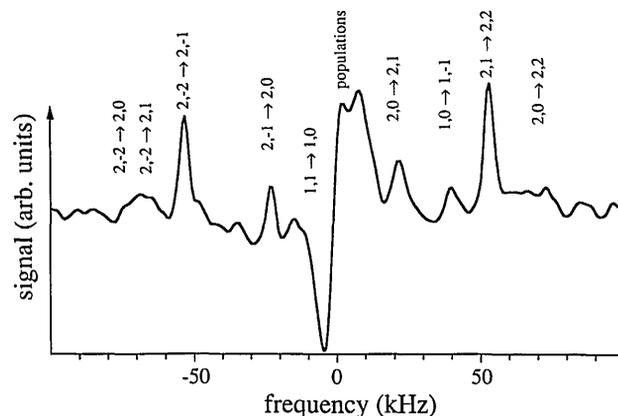


Fig. 2. Spectrum obtained by Fourier transformation of the experimental time-domain data from the Na ground state. Signal contributions from different multipole moments lead to different resonance lines. The numbers above the resonance lines indicate the assignment to the sublevel transitions in the form ( $F, m_F \leftrightarrow F', m_{F'}$ ).

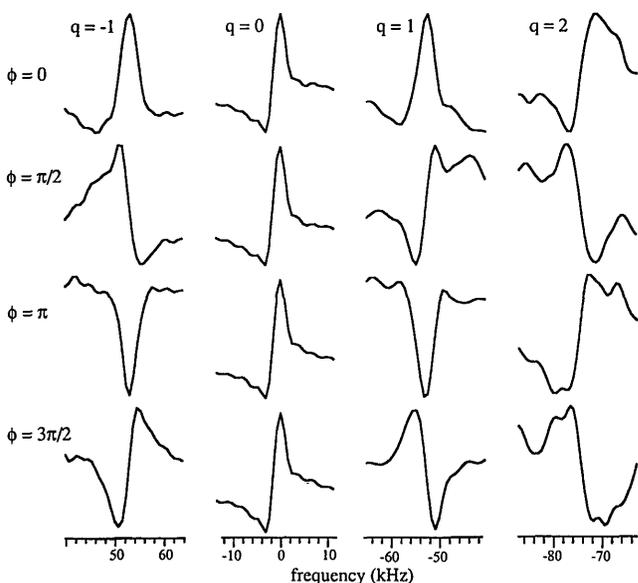


Fig. 3. Signal contributions from four different multipole moments compared for four modulation phases corresponding to different orientations of the atomic multipole moments.

to see an effect of the phase shift on the different resonances that is proportional to the order  $q = \Delta m_F$  of the corresponding multipole moments. Figure 3 shows the measured phase changes for four different resonance lines of the sublevel spectrum; each resonance line originates from a different multipole moment. The phase dependence is checked most easily for the resonance line labeled  $q = 0$ , which corresponds to axially symmetric sublevel polarization, i.e., to population differences. The shape of this resonance line is virtually identical in the four experiments, indicating that the corresponding atomic multipoles are invariant under rotations around the direction of the magnetic field.

The behavior of the line with  $q = 2$  is also relatively simple to analyze: the line shape is inverted whenever the modulation phase is shifted by  $\pi/2$ . The corresponding multipole moments obviously have a periodicity of  $\pi$ . The other resonance lines, labeled  $q = \pm 1$ , change the line shape from approximately absorptionlike to dispersionlike when the modulation phase is shifted by  $\pi/2$ . The periodicity of these multipole moments is apparently  $2\pi$ . These two resonance lines clearly show that the resulting phase change is sensitive not only to  $|q|$ , but also to the sign: the phase changes observed for these two lines have opposite signs.

The results obtained here show that it is possible to distinguish different orders of atomic multipole

moments by the different phases that they acquire during a rotation around the quantization axis. We demonstrated this technique experimentally for the Na ground state, using the  $D_1$  transition to prepare the multipoles by optical pumping and for monitoring them through the anisotropic optical properties of the medium. Here we have presented signal contributions that are due to multipole moments of order  $q = 0, \pm 1$ , and 2. We implemented the rotation of the density operator by using modulated optical pumping in a transverse magnetic field. Rotation of the density operator around the direction of the magnetic field could then be implemented by shifting the phase of the modulation. This method is not limited to Na or electronic ground states but can also be applied to excited states. This possibility should be attractive primarily in cases in which the different signal contributions are otherwise difficult to distinguish, such as in the case of severe spectral overlap or for the observation of weak signals that are obscured by stronger signals that are due to different multipole moments. Even experiments that achieve a separation of contributions from different multipole moments<sup>14</sup> may benefit from this technique, which may be used to enhance the signal-to-background ratio.

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