Fine Structure of Na $4d\ ^2D$ Using High-Resolution Two-Photon Spectroscopy*

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We have measured the fine-structure splitting of the $4d\ ^2D$ state of neutral sodium using high-resolution, cw, simultaneous two-photon excitation from the $3s\ ^2S$ ground state. Sodium atoms in an atomic beam were excited by a single-mode, cw, tunable dye laser, and the resonance was detected by monitoring the decay fluorescence from the excited state. The fine structure of the $4d\ ^2D$ state was well resolved and measured to be $\Delta_{fs} = 1025 \pm 6$ MHz.

We have measured the fine-structure splitting of the $4d\ ^2D$ state of neutral sodium using simultaneous two-photon excitation from the $3s\ ^2S$ ground state. The linewidth in the experiment (0.005 cm^{-1}) was sufficient to resolve fully the excited-state fine structure, as well as the ground-state hyperfine structure. We believe this to be the first high-resolution two-photon spectroscopy performed with visible radiation. It is also the first two-photon spectroscopy using photons from a cw laser.

Simultaneous two-photon excitation (as opposed to double resonance or stepwise excitation) was discussed theoretically by Göppert-Meyer[1] in 1929 and more recently by Gold.[2] Hughes and Grabner[3] first observed two-photon transitions, in an atomic beam at radio frequencies, in 1950, and two-photon spectroscopy has been performed with rf and infrared photons by Freund *et al.*[4] Two-photon excitation in atoms has been observed in the visible by Abella[5] and others,[6] but this work has all been done with pulsed lasers whose linewidth was typically 1 cm^{-1}, making precision spectroscopy impossible.

Two-photon spectroscopy offers many advantages for precision spectroscopy, especially in the light of recent advances in tunable cw laser technology: (1) access to S and D levels (inaccessible by single-photon dipole radiation from an S ground state), (2) extension of accessible frequency range for spectroscopy using tunable lasers by allowing tunable and fixed-frequency lasers to be combined, and (3) high resolution relative to stepwise-excitation experiments, since the excited-state linewidth does not depend on the width of the intermediate state. Some of these advantages have been pointed out before.[1,2,7]

We believe that two-photon spectroscopy offers a general technique for studying the structure of excited levels which is complementary to the method of quantum beats[8]: The former is preferable for structure whose spacing is several hundred megahertz or more, and the latter for more closely spaced structure.

Our experiment was performed on a supersonic beam of sodium atoms produced by an oven similar to Mattison's.[9] The tunable light source was manufactured by Spectra Physics and consists of a model-166 argon-ion laser which pumps a model-370 tunable dye laser fitted with a model-480 intracavity etalon for single-mode operation. The laser was operated at 17 274 cm^{-1}, exactly half of the $3S \rightarrow 4D$ transition energy. The laser light was focused by a 10-power microscope objective and intersected the atomic beam at a right angle. Decay fluorescence perpendicular to the plane of the intersecting laser light and atomic beam was collected by an $f/2$ lens and imaged onto a photomultiplier.

The search for the two-photon excitation and some initial measurements were made with an alkali density of roughly 10^{11} atoms/cm^3 and 165 mW of single-mode laser power. In subsequent measurements an interference filter was introduced to cut down background light. The filter had a 40-Å bandwidth and passed light from only the $3P \rightarrow 3S$ step in the decay from the $4D$ level. An aperture in front of the photomultiplier limited the region from which fluorescence could be observed so that the angular divergence of atoms whose fluorescence was detected was less than 0.01 rad.

Measurements of the detailed structure of the $3S \rightarrow 4D$ transition were made by sweeping the laser frequency at approximately 400 MHz/sec using the Spectra-Physics model-481 electronics package. The amplified output from the photomultiplier was passed through a 0.1-sec-time-constant RC filter and displayed on an oscilloscope synchronized with the frequency sweep of

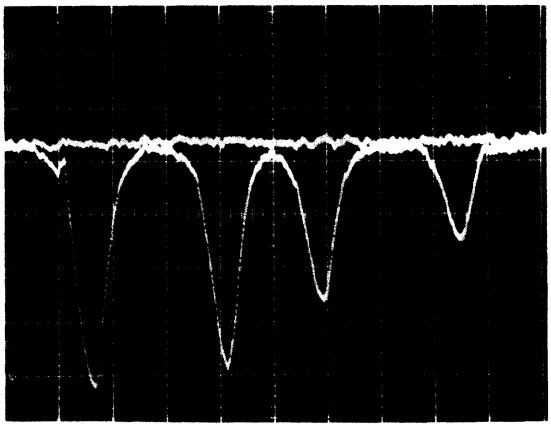

FIG. 1. The four components of the $^2S \rightarrow {}^2D$ transition in Na (peaks inverted) shown with frequency increasing approximately 200 MHz/division to the right.

the laser. A typical sweep is shown in Fig. 1. The largest peak corresponded to a signal of $\sim 10^5$ counts/sec above the background of $\sim 2\times 10^5$ counts/sec observed with the laser light blocked (horizontal trace in Fig. 1).

The dominant contribution to the 120-MHz linewidth observed in this experiment was the small size of the focused laser beam which limited the interaction time of the atoms with the field to roughly 2×10^{-9} sec. The laser linewidth (measured independently) was less than 25 MHz and broadening due to beam divergence was roughly 20 MHz. The 120-MHz linewidth in the present experimental setup prevented us from resolving the hfs in the 5S state, although we did observe the two-photon $3s\ ^2S \rightarrow 5s\ ^2S$ transition at 16 600 cm^{-1}.

The four peaks observed in Fig. 1 result from the fact that the $3s\ ^2S$ and $4d\ ^2D$ levels are both split into two components while the selection rules for two-photon absorption permit all four possible transitions. We attribute the four peaks to the following transitions (in order of increasing frequency): $^2S(F=2) \rightarrow {}^2D_{5/2}$, $^2S(F=2) \rightarrow {}^2D_{3/2}$, $^2S(F=1) \rightarrow {}^2D_{5/2}$, and $^2S(F=1) \rightarrow {}^2D_{3/2}$, where F is the quantum number for the total electronic and nuclear spin of sodium. No evidence of hyperfine splitting or quadrupole interaction in the excited state was observed.

An accurate determination of the fine-structure splitting in the $4d\ ^2D$ state was made by comparing the spacings of those components of the transition separated by the ^2D fine-structure energy difference Δ_{fs} with the spacing of those components separated by the well-known[10] hyperfine interaction energy of the ground state. This method eliminated errors arising from imperfect determination of the sweep amplitude and permitted a measurement of and correction for errors arising from uniform nonlinearity of the sweep as well. Our result, obtained by averaging 42 sweeps like the one shown in Fig. 1, is $\Delta_{fs} = 1025 \pm 6$ MHz. The error represents 3 standard deviations of the mean and exceeds our estimate of systematic errors. The best previous measurement of the fine-structure splitting in the 4D state was made by Meissner and Luft[11] in 1937 using interferometric techniques and an atomic beam. Their result, 1038 ± 50 MHz, agrees with our result.

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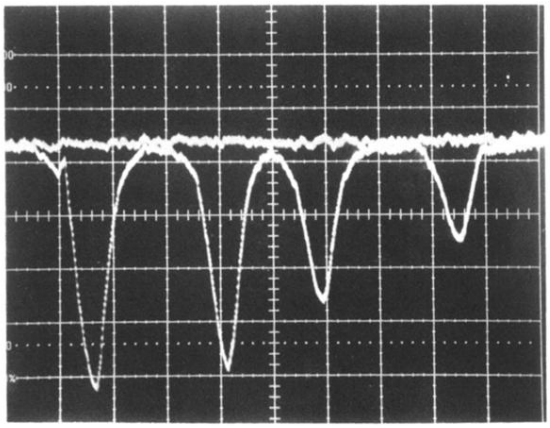

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